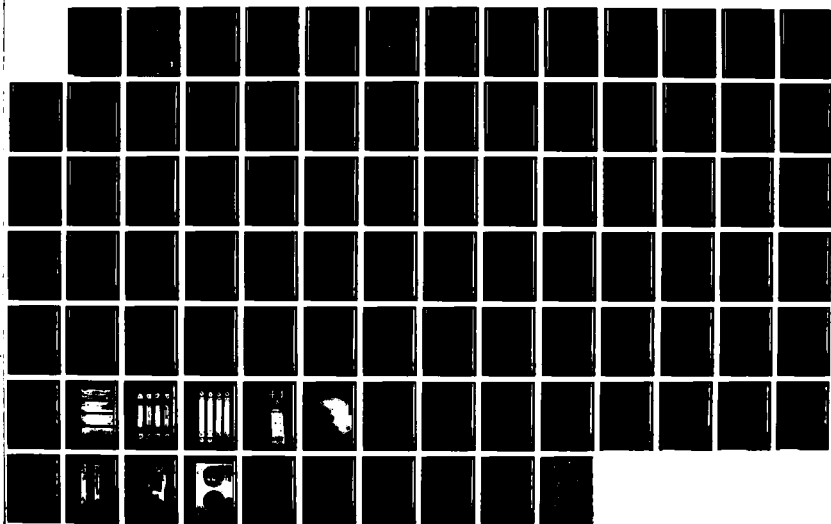AD-A191 027 DECONTAMINATION OF FAST REACTOR HULLS AND PROPERTIES OF   1/1
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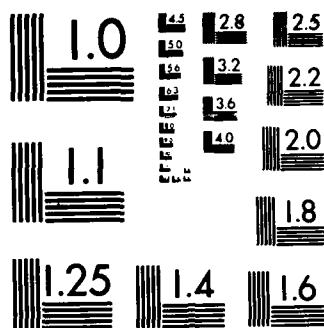   ESTABLISHMENT HARWELL (ENGLAND) P BIDDLE ET AL. OCT 86
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Decontamination of fast reactor hulls and properties of immobilised waste forms

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Decontamination of Fast Reactor Hulls and
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P. Biddle, P.E. Brown and J.H. Rees.

FINAL REPORT

Study performed under contract No. 312-83-2WAS. UK as
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Chemistry Division
Harwell Laboratory

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UNDERWATER APPLICATIONS OF THE
ULTRASONIC TIME-OF-FLIGHT DIFFRACTION TECHNIQUE
DRAFT

K. Newton, B.M. Hawker and A.P. Wein

ABSTRACT

This report summarises a programme of wor. carried out between 1982 and 1585 for the Harwell Offshore Inspection P and D Service. The main aims of the programme were to assess the potential of the ultrasonic time-of-flight diffraction (TOFD) technique for offshore applications, and then to develop, validate and quantify the performance of a complete TOFD system for the accurate sizing of defects in underwater welded node joints.

TOFD offers capabilities not possible with any other NDT technique available for use offshore. It complements the performance of ACPD for sizing surface-breaking defects, and it is the only available technique for accurate sizing of embedded defects or root cracks.

A State-of-the-Art Review was prepared at the beginning of the project which described the various Time- .-Flight techniques, considered the new factors encountered for offshore inspection and concluded that the techniques could in principle offer accurate sizing of defects in node welds.

Theoretical and experimental work considered the materials, coatings and geometries employed on offshore steel structures and established the feasibility of using TOFD techniques in this application.

Comprehensive validation trials for a wide range of geometries have indicated that a sizing accuracy of about 1 mm should generally be achievable.

A complete prototype TOFD system, interpretation software and inspection procedures for sizing known fatigue cracks in welded node joints underwater have been developed. Diver trials have indicated that scanning can be performed quickly and easily even by inexperienced divers, and data interpretation can be carried out on-site by a surface operator using a portable instrumentation system.

We believe that the programme has produced scanner designs, software and documentation of a sufficient standard to enable the TOFD technique for underwater crack sizing to be transferred to a service company, for routine application, with a minimum of development and training effort.

March 1986

ABSTRACT

The studies described in this Report have been carried out on five batches of hulls resulting from the reprocessing of fuel from the Prototype Fast Reactor at Dounreay. Direct immobilisation in cement and decontamination have been examined as possible routes for treating the hulls, and as a background to this work the activities on them have been characterised. The outcome of this project is described below.

Characterisation

>Five batches of hulls have been examined, corresponding to fuel with burn-ups in the range 0-7.3%. They have been examined visually, and found to be mostly either shiny (from lower breeder sections) or dark (from core or upper breeder regions). Photographic records of surface features and of the quality of the shear have been made.

The fissile activity on the hulls generally corresponded to less than 0.1% of the original fuel; the radionuclides were present on both the inner and outer surfaces. It is unlikely, however, that much of the activity present is due to undissolved fuel since deposition from dissolver liquors accounts for much of the activity present on the hulls.

Profile diagrams have been constructed to describe the levels of the various activities along selected hulls. Total α-activities varied between 4 and 200 $\mu Ci/g$. The principal fission products were: Ru-106 ($10 \cdot - 4 \times 10^5$ $\mu Ci/g$), Sb-125 ($4 \times 10^2 - 5 \times 10^3$ $\mu Ci/g$) and Cs-137 (20-500 $\mu Ci/g$). Other fission products measured include Cs-134, Ce-144 and Tc-99.

The principal activation products measured were Mn-54, Co-60 and Co-58. The value of the ratio of Mn-54:Co-60 can be used to indicate the position of the hull in the original fuel pin. C-14 levels of up to 5 $\mu Ci/g$ have been measured in the higher burn-up hulls.

Decontamination

A number of low temperature chemical techniques have been used to remove actinides and fission products from the hulls. The methods included: nitric acid at reflux temperature, nitric acid with ceric ions added, alkaline permanganate solution, and oxalic acid with hydrogen peroxide corrosion inhibitor. Irrespective of the method of decontamination or burn-up, the levels of α-activity could be reduced to less than 0.2 Ci/te of hulls. Thus decontamination might permit shallower, and therefore cheaper, disposal than would be the case for undecontaminated hulls.

The preferred route is that involving nitric acid alone, because it might be possible to recycle the decontamination liquor to the reprocessing plant, routeing the fission products and several of the actinides to the high level waste, while recovering the plutonium.

Immobilisation

10mm sections of hulls have been immobilised in two cements in which either sand or blast furnace slag was used to extend the ordinary Portland cement. Loadings of about 40g hull in 50mm right cylinders have been achieved. The activities released on leach testing these blocks have been compared with those from leaching bare hulls. The level of Cs-137 leached from hulls immobilised in either of the cement compositions was about an order of magnitude lower than that leached from the bare hulls. For Sr-90, the BFS/OPC matrix gave a tenfold better retention than the sand/OPC. Ru-106 and Sb-125 were leached from the sand/OPC but not from the BFS/OPC samples. The preliminary studies suggested that cement was a satisfactory matrix for immobilising hulls and that the BFS/OPC was the better formulation. Further evaluation of the waste forms was required.

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CONTENTS

LIST OF TABLES

LIST OF FIGURES

1. INTRODUCTION

In the head-end section of a reprocessing plant for irradiated oxide fuel, the fuel pins are sheared into 25-50 mm lengths which are routed to a basket in the dissolver. There they are treated with nitric acid which dissolves out the fuel, leaving the cladding residues, or hulls, as a solid waste product. In order to assess the options for the disposal of this waste it is necessary first to characterise the activities present both qualitatively and quantitatively. These activities arise in a number of ways, including:

(i) fuel which has been protected from acid attack by physical means, e.g. as a result of a poor shear;

(ii) atoms forced into the clad by fission recoil;

(iii) atoms which have diffused into the clad, particularly at grain boundaries;

(iv) activities deposited from the dissolver liquors; and

(v) activities produced by the neutron irradiation of the cladding atoms.

The most favoured disposal route for solid wastes in the CEC is by burial in geological formations following immobilisation in a suitable matrix. The depth required for burial may be dependent on the quantity of very long-lived isotopes the waste contains, and thus a preliminary decontamination step to remove such activities from the hulls might lead to shallower and therefore less costly disposal.

The present work is concerned with stainless steel hulls originating from the reprocessing of fuel from the Prototype Fast Reactor (PFR) at Dounreay (U.K.). The studies were jointly funded by the U.K. Department of the Environment and the Commission of the European Communities. The objectives were to characterise the activities present on genuine hulls (particularly long-lived isotopes that could be important in disposal.), to examine the possibility of decontaminating them by simple, low temperature, chemical techniques, and to study the immobilisation of the undecontaminated hulls in cement matrices, with evaluation of the waste forms mainly by leach testing. The matrices studied have been used in the U.K. waste treatment programme, where cement is the leading option for immobilisation of non-heat generating wastes.

The results obtained should be of value in formulating policies for the management of hulls arising from future commercial fast reactors.

A number of procedures were developed by Jenkins and coworkers[1] for characterising the activities on hulls from thermal reactors. These included total fissile measurements by delayed neutron counting, activity distribution measurements by fission track and α-autoradiography, α-spectrometry of surfaces and depth penetration measurements, as well as standard radio-analytical procedures on solutions resulting from the dissolution of the hulls. Some of these techniques have been employed in the current work and profiles of the activities along various hulls constructed.

The decontamination of stainless steel is a very wide field which has been included in a number of reviews[2,3,4]. Much of this work was concerned with the decontamination of coolant systems in water reactors, but the results served as a useful guide for a preliminary experimental study of the decontamination of stainless steel hulls using samples from experimental pins in the Dounreay Fast Reactor[5]. The findings from this study provided a framework for the decontamination of the PFR hulls.

The conditions and techniques used in the immobilisation and leach testing were based on earlier work at Harwell with fuel cladding derived from thermal reactor systems.

2. ACTIVITIES ASSOCIATED WITH HULLS

There are three types of activity associated with hulls: α-emitters, fission products and activation products. The latter arise from the neutron irradiation of the cladding whereas the two former types originate from the fuel.

2.1 Predominant Activities

The principal α-emitters found in earlier studies were Pu-239, Pu-238, Pu-240, Am-241, Cm-242 and Cm-244[1,6].

The most prolific fission product activity found was Ru-106 followed by Sb-125, Cs-137, Cs-134, Sr-90 and Ce-144, with smaller quantities of Eu-155, Eu-154 and Ag-110m.

The main activation products measured were Mn-54, Co-60 and Co-58.

All the above isotopes, apart from those of Pu and Am, have half lives of 30 years or less, and can be considered short lived in terms of disposal.

2.2 Other Activities

There are also a number of low-energy fission and activation products which are long lived, and consequently could be important in considerations of the long-term movement of nuclides from repositories. They include Tc-99, I-129 and C-14. Np-237 is another isotope which needs to be examined in this context, as it is potentially one of the more mobile actinides. As a first step, calculations were carried out to estimate the quantities of these isotopes likely to be found on the hulls.

2.2.1 Technetium-99

Calculations using the FISPIN computer code indicated that for core region fuel subjected to 7.2% burn-up, the Ru-106/Tc-99 activity ratio was approximately 7.5×10^3 after four years' cooling[7]. PFR hulls had previously been observed to contain 0.2 to 60 mCi of Ru-106 per gram of hull (with a mean value of 12 mCi/g) after a cooling period of four years[6]. On this basis an estimate was made of the range of Tc-99 activities which might be encountered in PFR hulls, namely 0.03 to 8 µCi of Tc-99 per gram of hull. This represents 1.5 to 500 µg Tc-99/g hull, with a mean value around 100 µg/g. Measurement of such levels should be feasible, and the isotope was accordingly included in the programme.

2.2.2 Iodine-129

The relative fission yields of I-129 and Tc-99 in PFR fuel were obtained from FISPIN calculations and the data combined with the results of the Tc-99 assessment. The estimated range of I-129 activity was 0.04 - 12 nCi per g hull. This is likely to be a considerable overestimate because the higher solubility and volatility of iodine in the dissolver relative to technetium should be taken into account. The abundance of I-129 on PFR hulls is likely to be far too low for direct counting and a separation/concentration procedure would probably be very complex. This isotope was therefore not included in the present programme.

2.2.3 Neptunium-237

Comparing the relative activities of Np-237 to those of Pu-238 and (Pu-239+Pu-240) obtained by FISPIN computations with the quantities of the Pu isotopes previously measured experimentally[6], it was concluded that the level of Np-237 on the hulls was unlikely to exceed 550 pCi/g hull; this is too low to measure using normal radiometric or chemical analysis.

2.2.4 Carbon-14

C-14 in PFR hulls is predominantly produced by an (n,p) reaction involving the N-14 impurity in the stainless steel cladding. Assuming a nitrogen level of ~ 200 ppm in the steel, the rate of C-14 production would be about 20 Ci per GW(e) year of power generation[8]. For a moderate fuel burn-up of 7.2%, some 304 sub-assemblies would be required to generate 1 GW(e) year, leading to a C-14 activity of 66 mCi per sub-assembly. The fuel cans in each sub-assembly have a mass of approximately 40 kg, representing about 35% by weight of the total cladding (wrapper plus spacer grids plus fuel cans). The irradiated hulls from one sub-assembly would therefore be expected to contain 23 mCi of C-14, which represents a specific activity of ~ 0.6 µCi C-14/g hull. It was thought that measurement of such levels in the hulls should be feasible, and the isotope was therefore included in the experimental programme.

3. THE PFR FUEL PIN

The PFR pins from which the hulls originate comprise four main sections: the core, the upper and lower axial breeders, and the gas plenum (Fig. 1). Molybdenum wire spacers housed in a stainless steel body are situated between the plenum and the lower breeder and between the lower breeder and the core. The position of these spacers can be seen on the outside of the cladding as a crimped section (see Figure 2(b)), and this allows identification of the originating position of some of the hulls.

4. THE HULL SAMPLES USED

4.1 Post-Dissolver Treatment

After completion of the dissolution of fuel from the pin sections, the basket of hulls was lifted up in the dissolver and the liquors allowed to drain off. The basket was then transferred to a washing vessel where the next charge of acid en route to the dissolver was passed over the hulls at ambient temperature. After draining, the hulls were dried by hot air. Thus a further source of activity on the hulls, via from the evaporation of the wash liquor residues, was introduced. On receipt the hulls were generally dusty with a lot of loose contamination, much which probably originated from this drying step.

4.2 Details of Hull Batches

Five batches of hulls with burn-ups ranging from 0 to 7.3% and cooling times of up to 6 years were examined. The hulls were generally 25-75 mm long.

The batches were designated A, B, C, D and E and details relating to the fuel they had contained are given in Table 1. Batch B contained hulls from two sub-assemblies of different burn-ups and was not used in the characterisation and decontamination work. Batch D consisted of hulls from pins which had been fuelled but had not been irradiated. They were, however, sheared in the active facility and the fuel removed in the same dissolver that had been used for irradiated fuel. Batch E contained some pin end-piece hulls which were up to 110 mm long.

4.3 Visual and Photographic Inspection

Each batch of hulls was examined visually and a number (usually ~6) selected for further study. The basis of selection was to obtain mainly typical samples together with a few which exhibited special features. From their appearance, the hulls could be separated into two types, black and shiny (Fig. 3). A number of the shiny hulls contained crimped sections, indicating the presence of a spacer. This showed that these hulls originated from the lower axial breeder regions of the pins. Observations on whole pins at Dounreay[9] have confirmed that around the lower breeder region the pins appear shiny whereas in the core and upper breeder regions they are dark and matt.

Each of the selected hulls was photographed from both end and side viewpoints (examples are shown in Figs. 3 and 4). The end views showed the quality of the shear which was generally good, with open ends affording easy access for the dissolver acid to the inside of the hulls. The side views (see Fig. 2) showed surface features along the length of the hulls. Some of the surfaces were clean whereas others were coated with oxide films of varying thicknesses which produced a mottled effect. On some hulls the presence of particulate material was noted. The oxide films are formed by interaction of the clad with the low levels of oxygen present in the coolant under the conditions of the irradiation and provide additional sites for the absorption of fission products and other activities. Oxide films will also be present on the internal surfaces of the hulls due to interaction between the cladding and the fuel.

Another feature shown in the side views is the location of spacers; the structure of a spacer is shown in Fig. 7. In one case where the crimped section was very close to the end of the hull, the shear had actually gone through the spacer body and a cross section of this can be seen, somewhat distorted, in the end view of hull number 4 in Fig. 3.

4.4 Cutting of Hulls

The radiation levels from the hulls were very high. The facilities available for the characterisation and decontamination work consisted of a number of shielded fume hoods, and in order to be able to use these facilities without incurring high radiation doses to the operator, it was necessary to limit the size of hull samples to rings about 1 mm wide. This is similar to the 1-2 mm rings used by Jenkins et al[1] in their work on thermal hulls.

About ten 1 mm rings were cut from a defined end of each of the selected hulls, and the individual rings were separately packaged so that their exact location along the length of the hull was known.

The optimum size of hull slice for packing into the moulds for cement immobilisation on a small scale had been found from inactive trials to be about 10 mm, so in order to provide sufficient material to make about 11 samples of immobilised waste the bulk of the material from each batch of hulls was cut to this length.

5. EXPERIMENTAL

5.1 Characterisation

5.1.1 Total Fissile Measurements

A weighed ring of hull was sealed into a polythene tube and irradiated briefly for a known time in a reactor. After a short, measured cooling time, the delayed neutrons emitted were counted during a known time interval. From the experimental data it was possible to calculate the amount of fissile material but not to differentiate the contributing isotopes. The results were normally expressed as though all the fissile material was Pu-239.

5.1.2 Fission Track and α-Autoradiography

In Jenkins' work[1] fission track autoradiography had shown that fissile materials were present in significant quantities on both the inner and outer surfaces of the hulls. Since the only time the outer surfaces came into contact with fissile material was in the dissolver liquors, these results pointed to deposition having occurred. Owing to .he higher levels of activity associated with the PFR hulls, it was not possible to use this technique with the handling facilities that were available. A few measurements using α-autoradiography, however, confirmed that the fast reactor hulls also contained actinides on both inner and outer surfaces, that they were restricted to the top 2-3 μm, and that the amounts were very variable.

5.1.3 Dissolution Experiments

The hulls were dissolved by refluxing with 5M nitric acid - 2M hydrochloric acid for about a half to one hour.

The principal fission and activation products in the solutions were determined by γ-spectrometry using a lithium-drifted germanium detector connected to an Intertechnique IN90 Programmable Multichannel Analyser.

Sr-90 was determined by β-counting following a radiochemical separation[10].

The gross α-content of the solutions was found by evaporating small aliquots to dryness on stainless steel discs, briefly flaming them, and measuring the α-activity with a scintillation counter. To obtain information about the component isotopes, a solvent extraction separation was carried out. The Pu in the solution was conditioned to the four valent state by adding a few crystals of sodium nitrite and the solution contacted with an equal volume of 10% tri-n-octylamine in xylene. Under these conditions the majority of the Pu extracted into the solvent phase whilst the Am and Cm remained in the aqueous. α-Spectrometry was carried out on each phase.

The plutonium isotopic composition was obtained by mass spectrometry on the hull dissolution solution after an ion-exchange clean-up to remove the steel components and fission products.

5.1.4 Tc-99 Determination

The hull was dissolved in 3M sulphuric acid and conditioned with hydrogen peroxide to ensure that the Tc was present as the pertechnetate. The Tc was separated from the other fission and activation products by first treating the hot solution with tetraphenylarsonium ions in the presence of a perchloric acid carrier[11]. On cooling, the tetraphenylarsonium pertechnetate and perchlorate cocrystallised almost quantitatively. The crystals were filtered off, dissolved in ethanol and passed through an anion exchange column. The tetraphenylarsonium cation was removed by washing with ethanol and the technetium then eluted with perchloric acid and reduced to Tc(V) with ascorbic acid in the presence of thiocyanate. The red Tc thiocyanate complex was extracted with butyl acetate and analysed by spectrophotometry[12], using the absorption at 516 nm. The limit of detection was about 1 µg Tc.

5.1.5 C-14 Determination

The hulls were dissolved by refluxing in an oxidising acid medium which released the C-14 as a mixture of carbon dioxide, carbon monoxide and simple hydrocarbons. These gaseous products were transported by a nitrogen stream to a catalyst column where the carbon monoxide and hydrocarbons were converted to carbon dioxide. The catalyst used was 300-850 µm cupric oxide heated to 400-425°C. The cupric oxide was prepared by precipitation from hot cupric sulphate solution; according to Kabat[13] this route produces the most effective form of catalyst. After leaving the catalyst column the gas stream was passed through a sodium hydroxide absorption train to remove the carbon dioxide. The alkaline solution was then analysed for carbon-14 by liquid scintillation counting using an Instagel or Picofluor-30 cocktail.

A number of preliminary tests on the experimental equipment were carried out. These showed that no carry-over of fission product radioactivity occurred during the dissolution of an active hull, gave information on the likely extent of acid carry-over during a run, and established that no measurable "blanks" would be produced. The recoverability of C-14 from the system was examined by adding C-14 labelled sodium carbonate to the acid in the dissolver and conducting a run in the usual way. Initially recoveries no better than 70% were

obtained, but these rose to a consistent 90% when the cupric oxide was
omitted, indicating that contrary to Kabat's findings, some absorption
of C-14 on to the copper oxide was taking place. The remaining 10% of
the C-14 can be accounted for in terms of counting errors and absorption
on the surfaces of the equipment. As a result of these findings the
apparatus was modified for the active runs by the insertion of a second
sodium hydroxide absorption train in front of the catalyst column.

5.2 Decontamination

The decontamination procedure consisted of treating the weighed hull
slice under the specified conditions, followed by retrieval, removal of the
excess liquid by blotting on filter paper, and reweighing after each stage.
Finally the hull was dissolved in a nitric-hydrochloric acid mixture and the
solution analysed in order to establish the residual, and hence the total
activity, associated with the original hull.

Earlier work[5,6] had indicated that high levels of decontamination
could be achieved using four different reagents:

(1) Nitric Acid

The standard conditions using this reagent were 9 molar acid at
reflux. The effect of the number of contacts, the duration of the
contacts and the variation of acid concentration and temperature were
studied.

(2) Nitric Acid in the Presence of Ceric Ions

Two molar nitric acid containing 0.1 molar ceric ions were used in
single one hour contactings at 90-95°C.

(3) Alkaline Permanganate

This was a two-stage process involving firstly a treatment at
reflux with 3% potassium permanganate – 10% sodium hydroxide followed by
a wash based on the CITROX formula consisting of 2.5% oxalic acid and 5%
diammonium hydrogen citrate at 85°C.

(4) Oxalic Acid

Oxalic acid was used both on its own and in the presence of a
hydrogen peroxide corrosion inhibitor. Treatments were normally carried
out at reflux and involved one or more contactings of 2-6 hours'
duration. The effect of varying the oxalic acid concentration was
examined.

5.3 Immobilisation

5.3.1 Inactive Trials

The technique adopted for preparing the immobilised products was established inactively before the equipment was installed for remote application in a shielded facility. It involved vibrocompacting a weighed quantity of the hulls in a plastic mould with the appropriate amount of cement grout (Fig. 5), which comprised either sand/ordinary Portland cement (OPC)/water or blast furnace slag (BFS)/OPC/water in the ratios 3:1:0.48 and 3:1:1.28 respectively by weight. The size of the immobilised product was chosen to be a 50mm right cylinder (Fig. 6) to facilitate handling and storage of the active samples during leach testing. To obtain an adequate loading of the hulls their lengths were reduced from a nominal 50 mm to 10 mm.

After vibrocompaction the cement samples were kept in a moist atmosphere for 48 hours before demoulding and curing. Samples containing sand/OPC were cured for 28 days and those containing BFS/OPC for 90 days, both at ambient temperature, prior to leach testing. This was to ensure that the samples had attained a comparable strength and microstructure.

5.3.2 Active Studies

The active, immobilised samples were made in a shielded facility according to the optimised conditions established from inactive trials. Eleven immobilised samples containing sections of irradiated PFR hulls were made (see Table 2). In one case some inactive material was included to make up the weight. Nine of the samples were leach tested and two were stored as archive samples pending further examination.

A sample of the waste to be immobilised was analysed radiochemically and it was assumed that this was representative of the complete batch of waste and hence of the total activity (A_o) that was incorporated in the final product. Leachates were subsequently analysed for all the detectable radionuclides. The Cs, Ru and Sb activities were determined by γ-spectrometry, the Sr by β-counting following a radiochemical separation, and the actinides by scintillation counting. Bare hulls were leach tested with (i) distilled water, (ii) water that had previously been equilibrated with sand/OPC matrix, and (iii) water that had earlier been equilibrated with BFS/OPC matrix, to ascertain the effectiveness of immobilisation.

5.3.3 Leach Testing

The conditions and methods of leaching were a variant of those recommended by the International Standards Organisation test - ISO 6961 (1980). The immobilised waste form was suspended in 295 ml of distilled water contained in a lidded polypropylene pot. The water was changed after a one day contacting and four further changes were made at intervals during the next 13 days. This was followed by four contacts of 7 days and two or three of 28 days, all at ambient temperature. In each case, the water after removal from the waste was acidified with 5 ml nitric acid prior to analysis to keep the leached activities in solution. In the case of bare hulls, ten 10 cm lengths of hull were suspended on a plastic mesh within the polypropylene container.

It was noted that in some cases the γ-radiation emitted from the immobilised waste forms was sufficient to cause embrittlement of the plastic container within about 28 days. The use of stainless steel containers in future studies is recommended.

6. RESULTS AND DISCUSSION

6.1 Characterisation

6.1.1 Total Fissile Content

The total fissile contents of rings cut from hulls of each of the batches are given in Table 3. The values assume all the fissile material to be Pu-239. There were considerable variations in the results, with some of the highest values being found in hulls where it was least expected, for example, on unirradiated hulls and on ones from around the lower breeder regions. This suggested that deposition was making a significant contribution to the fissile material on the hulls. The variability of fissile contents of fast reactor hulls was previously reported by Jenkins[1,14] in connection with hulls from the Dounreay Fast Reactor.

As the density and Pu content of the original fuel were known, the fissile contents could be expressed as percentages of the initial fuel associated with the particular hull slice. The values obtained were generally less than 0.1%. These are not, however, realistic measures of the amount of undissolved fuel associated with the hulls due to the apparent importance of deposition.

6.1.2 α-Emitters - Constituent Isotopes

α-Spectrometry was used to analyse the phases obtained from the solvent extraction treatment of the dissolved hull solutions (see Section 5.1.3).

The organic phases showed two peaks, one due to Pu-239 + Pu-240, the other to Pu-238 + Am-241. Because of the similarity of the α-energies of the constituent isotopes, these peaks could not be resolved further, but since the partition coefficient for Am-241 was very low indeed under the conditions of the extraction, the second peak was attributed solely to Pu-238. The aqueous phases produced peaks corresponding to Am-241+Pu-238, Cm-242 and Cm-243+Cm-244, together with a small peak due to unextracted Pu-239+Pu-240. Difficulties were encountered in analysing these aqueous phases owing to the high salt content that originated in the stainless steel. This meant that only small aliquots of the solution could be mounted for counting, and even then the sources were sufficiently thick to give rise to some self-absorption which resulted in a loss of resolution and peak shifting. The small Pu-238 contribution to the Am-241 peak could be corrected for by using the ratio (Pu-239+Pu-240):Pu-238 obtained from the organic phase results. The curium peaks were generally small and the associated counting statistics poor, even on very long counts. Because the principal isotope Cm-242 was a relatively short half life (163 days), it is difficult to correct the activity back to a reference date (e.g. the completion of the irradiation) without introducing significant errors. From a study of FISPIN data, Cm-244 should make the major contribution to the other Cm peak.

The results presented in Table 4 show the percentages of the individual isotopes in each phase on an activity basis. The columns labelled "OTHER" normally refer to small unidentified peaks but in a few cases where it was not possible to obtain data concerning the individual curium isotopes, the gross Cm figures are included under this heading.

By combining the percentage compositions and the gross α-counts of each phase, the overall percentage composition of the hull solutions can be computed. This is shown on the right hand side of Table 4. The data was obtained at three different dates over a seven month period, and because of its short half life the amount of Cm-242 altered considerably during this period, thereby altering significantly the percentage composition of the phases. In order to permit a comparison to be made, the Cm-242 activities were corrected to 16th February 1984 and the resulting compositions calculated. The corrected figures are given in Table 5 and are compared with percentage compositions obtained from

FISPIN data for the fuel. In view of the uncertainties involved a reasonable correlation appears to exist. This is also shown by the ratios of the isotopes Am-241:Pu-238: (Pu-239+Pu-240) given in Table 5.

The actinide composition of the fuel will vary according to its position along the fuel pin. When FISPIN calculations for the core region and the axial breeder regions are compared it can be shown that the weight ratio of (Pu-239+Pu-240:Am-241) is approximately 30 for the core regions but ~ 5.3×10^4 for the breeder regions. Values for this ratio computed from the data obtained in the present work varied from 70 to 150 irrespective of the position of the hull in the fuel pin.

These findings again suggest that the activity on the hulls arises largely by deposition from the dissolver liquors.

The isotopic composition of the plutonium found on some of the hulls is reported in Table 6. Uranium was also sought but the levels were too low to be detected.

6.1.3 Activation Products

The principal activation products detected by γ-spectrometry were Mn-54, Co-60 and Co-58. The Co-58, because of its short half-life (70 days), had largely decayed by the time of measurement and hence only small peaks were obtained. It was generally found on the higher burn-up hulls. The relative quantities of Mn-54 and Co-60, corrected to the time of completion of irradiation, were found to vary according to the position of the hull along the fuel pin. For the shiny hulls originating from around the lower axial breeder regions, the ratio Mn-54:Co-60 was low (<5), for black hulls from the core region values of 75-110 were observed, and for a hull from the upper breeder section (identified by the presence of a fuel retainer in the hull) a value of about 1 was obtained. Thus the ratio Mn-54:Co-60 can be regarded as an indicator of the location of the hull in the fuel pin. Reference to the profile diagrams (see Section 6.1.4) showed that it was the levels of Mn-54 that changed more than those of Co-60 along the lengths of the fuel pins. Table 7 gives the levels of activation products for a number of hulls which include one containing a spacer. This hull (A/5) was sectioned right across the spacer body and from the measurements and visual observations of the geometry of the individual slices, it was possible to ascertain the orientation of the hull in the original fuel

pin. A diagram of a spacer is included in Fig. 7. In the fuel pin, the end containing the molybdenum wire disc is uppermost. At the lower end of the hull just below the spacer no Mn-54 could be detected. Around the position of the crimp, values of 0.08 and 0.10 were found for Mn-54:Co-60. The values rose to 0.13 at the position of the molybdenum wire disc, 0.48 just above the spacer and 0.54 at the upper end of the hull. The sample was thought to have originated from between the lower axial breeder and the plenum regions.

The other activation product measured was carbon-14. On 4.1% burn-up hulls the activity levels of this isotope increased from 0.06 to 0.44 μ Ci/g with increasing neutron irradiation as indicated by the Mn-54:Co-60 ratio (Table 8). The 7.3% burn-up hulls followed a similar pattern with values ranging from 0.15 to 1.0 μ Ci/g, although higher values (up to 5 μCi/g) were found on two of the hulls with intermediate Mn-54:Co-60 ratios. The results obtained were in reasonable accord with the figure of 0.6 μCi/g predicted from calculations (see Section 2.2.4).

6.1.4 Fission Products and Activity Profiles

The combined analytical results from dissolution and decontamination experiments provided a considerable amount of data relating to the activity content of sections from a number of hulls. Since the relative positions of the various sections along a given hull were known, it was possible to construct an activity profile diagram for each hull examined, and thus provide a simple method for comparing different hulls. The profile diagrams (Figs. 7-10) show the activity in μCi/g hull decay-corrected to the date of completion of irradiation, plotted vertically on a log scale, and the location of the hull section relative to the hull end, shown on the horizontal axis. The black squares correspond to hull material lost during cutting. The dotted lines indicate the presence of sections for which data is not available. The diagrams show that the range of activity levels for the different isotopes varies considerably - 6 orders of magnitude. The levels of the activation products Mn-54 and Co-60 are fairly constant along a given hull, whereas the other activities show greater variations and can change considerably over quite short distances. In spite of this the various isotopes often follow similar patterns. In general the order of the activities is Ru-106>>Sb-125>Cs-137>total α. Sr-90, where data is available, normally lies between Cs-137 and total α. Table 9 gives numerical values for the average activities on 12 of the hulls examined.

The possibility of explaining the wide variation of activity levels along the hulls in terms of physical losses from the surface oxide film during the cutting of the 1 mm slices was considered. It was noted in earlier work however, that when slices o the same size had been cut under identical conditions from both Zircaloy and stainless steel thermal hulls, the activities along these hulls were reasonably uniform. Samples previously cut from experimental hulls from the Dounreay Fast Reactor had shown some variations though not quite as large as those on hulls from PFR. The significant variations appeared to be confined to fast reactor hulls and may be due to the plate out of very small discrete particles of dissolver insoluble material which is found in much greater quantities in solutions of irradiated fast reactor fuel than in dissolved thermal reactor fuel. The experimental observations are consistent with this, since the particulate material is normally rich in ruthenium and this is the nuclide that has shown the greatest variation along the hulls. In addition the presence of particulate material adhering to the surface of some of the hulls has been observed (see for example Fig. 2d).

A number of 10 mm sections of the batch 1 hulls were dissolved in acid in the hot cells and the solutions analysed. With these larger samples any effects of "cutting losses" would be smaller. When the levels of the activities were compared with those from 1 mm slices from hulls of the same batch, they were found to be very similar. The comparison for the principal fission products activities is made pictorially in Fig. 11. The first column in the figure contains data for the 10 mm slices, the second for the 1 mm slices and since the number of results is rather small, the third column gives data for similar 1 mm slices from the same batch but obtained outside of the present contract. It can be seen that the activities of the different isotopes fall within well defined bands with only a very small amount of overlap. The width of the bands was slightly wider at the 1 mm end but the extension was upwards rather than downwards. It seems unlikely therefore, that significant losses of activity were encountered during the cutting operations, other than that of loose activity arising from the drying out of dissolver liquor residues which would be partly lost anyway in general handling and cannot really be considered as part of

the activity of the hull. These findings were later confirmed when the analytical data from the complete hull decontaminations (see Section 6.2.7) were calculated. The hulls involved had not been cut at all, but the activity levels still fell within the same bands (see fourth column of Fig. 11). Corresponding plots of the activation product activity levels found in this series of experiments are given in Fig.12, which shows clearly the difference between the black and the shiny hulls (no 'in-between' hulls were involved). The width of the activation product activity bands are smaller than those of the fission products : this is attributed to their being integral to the hulls and not containing significant contributions from deposition.

The technetium-99 content of a high burn-up core hull (C2/D) was determined by dissolving the hull in dilute sulphuric acid and analysing the solution by the procedure described in Section 5.1.4. The value obtained, 40 μg Tc/g hull, was about the middle of the calculated range (see Section 2.2.1).

6.1.5 Activities Associated with End-Pieces

There were five pin end-pieces among the batch E hulls. These will have been top end caps since the plenum ends of the pins are not normally routed via the dissolver. They varied in length from 30-110mm and some contained plugs of solid material which were identified by electron microprobe analysis as UO_2. These plugs were located near the fuel retainer (see Fig.1), and must have been part of the upper breeder fuel which had been incompletely removed in the dissolver, because the closed end of the hull had restricted the access of acid to the interior. One millimetre rings were cut from each end of two of these hulls as shown in Fig. 13. They were dissolved in nitric-hydrochloric acid and the solutions analysed. The activity levels are shown in Fig. 14 and Table 10. For both hulls the Mn-54:Co-60 ratios increased with the distance of the rings from the closed end. This was not surprising as the neutron dose received would also be increasing. Comparing the profiles for the α-activities with those from ordinary hulls of the same batch showed slightly higher levels associated with these end pieces. This was presumably due to deposition since some of the rings were from clad which had not contained fuel. Data from a single ring of hull E1/13D taken 28 mm from the closed end showed generally lower levels of activity than found on the other two hulls at comparable positions.

6.1.6 Activities on Unirradiated Hulls

The activity on these hulls must have arisen from deposition in the dissolver, and its measurement provided an indication of the amount of activity a normal hull could acquire in this way from the dissolver liquors, bearing in mind that the surface of the other hulls had been modified by oxide film formation during the irradiation. Table 11 compares the activities found on unirradiated hulls with some typical mean values for hulls from batches A and C which passed through the dissolver during the same reprocessing campaign. In each case the fission product activities of the unirradiated hulls were lower than the irradiated ones (particularly for Cs-137), but the α-activities were about a factor of 2 greater. In view of the large variation of activities that has been observed on various hulls this may not be very significant. An attempt was made to compare the ratios of the principal fission product activities on the various hulls with those expected to be present in the dissolver liquors as calculated from FISPIN data. In general, the unirradiated hulls had ratios of activities which were fairly similar to those found on the irradiated hulls, but differed from the predicted values for the dissolver liquors, in that they had lower proportions of Ru-106, and much lower proportions of Cs-137, relative to Sb-125. This suggested that deposition of various isotopes occurred to different extents.

6.2 Decontamination

The main purpose of decontaminating the hulls is to remove those long-lived isotopes (mainly the α-emitters) that could potentially lead to problems in the long term when the waste has been disposed to an underground repository.

Two techniques were considered in earlier studies on the decontamination of the hulls, namely treatment with hot solutions and the use of ultrasonic radiation at ambient temperature. Both of these techniques gave satisfactory levels of decontamination under suitable conditions. The ultrasonic treatment, however, tended to remove significant amounts of the activity in particulate form rather than in solution; another disadvantage was that the method was likely to be difficult to scale up effectively. The use of ultrasonics was therefore not pursued further.

The bulk of the decontamination work concentrated on the four systems identified in Section 5.2.

6.2.1 N⸱⸱⸱⸱⸱ Acid at Reflux

Nitric acid is the most attractive decontaminant since it has the potential to produce low salt content wastes which are easy to deal with in a reprocessing plant.

The decontamination mechanism involves the dissolution of the oxide layer including any residual fuel particles. Earlier work suggested that repeated reflux with 9M acid gave the best results. Table 12 shows how the activity was removed during three successive six-hour contactings with fresh acid for hulls of two different burn-ups. In each case most of the α-activity removal occurred during the first contacting, the second and third contactings being effectively polishing stages. For fission products, lower removals were achieved during the first stage and there were quite significant further removals during the second stage.

Table 12 shows the overall percentage removals for a number of 5.6 and 7.3% burn-up hulls treated under the conditions described above. The hulls examined originated from a wide range of different positions along the fuel pins as indicated by the $Mn-54:Co-60$ ratios. In general, high levels of removal of α-emitters, of $Sb-125$, and of the minor fission product constituents were achieved, whereas the removals of $Ru-106$ and $Cs-137$ were more variable and tended to be lower. Reducing the temperature at which the contactings were made to 60°C resulted in a dramatic lowering in the decontamination performance. On the other hand, when the temperature was kept at reflux and the contact times were reduced to two hours, the removals of α-emitters were hardly affected and the fission product removals only slightly reduced. Lowering the contact time futher to one hour did not produce any great change in the decontamination efficiency. The main effect of using shorter contact times was to decrease the weight losses from the hull slices in most cases.

The weight losses need to be kept to a minimum, particularly if the acid is to be recycled through the reprocessing plant, since the presence of ferric ions might enhance dissolver corrosion and could produce an unwelcome increase in the salt content of the highly active waste liquors.

The final column in Table 13 gives data for the thickness of hull removed during the decontaminations. These values were calculated assuming a uniform loss of hull material from both inner and outer surfaces and from the ends. Since this is not strictly true, the values can only be regarded as an "order of magnitude" guide.

The effect of reducing the nitric acid concentration to 6M was also studied. The reduced level of decontamination was not very marked but was more significant for the fission products than the α-emitters.

Since percentage removal can be a misleading parameter in decontamination studies where variable amounts of initial activity are involved, the initial and residual activities in all the runs are also given (Table 14). This aspect is discussed further in Section 6.2.6.

6.2.2 Nitric Acid in the Presence of Ceric Ions

The use of this reagent as a decontaminant has been described by Partridge et al[15]. The ceric ion oxidises iron and chromium in the steel's surface causing them to dissolve in the nitric acid. It therefore decontaminates by removal of the surface layer, and one of the chief disadvantages of the reagent is that it cannot be used in normal stainless steel vessels.

Single one-hour contacts at about 90°C were used with 2M nitric acid containing 0.1M ceric ions. Under these conditions very good decontamination from both α-emitters and fission products (except for Ru-106) was achieved, the results (Table 15) being similar to those obtained over much longer periods with stronger nitric acid on its own. The weight losses from the hulls were however quite high, probably partly due to the aggressive nature of the reagent, and partly to attack through the unprotected cut ends of the hull slices.

Reducing the ceric ion concentration to 0.05M, whilst keeping the other conditions unchanged, led to reduced decontamination efficiency but lower weight losses. The efficiency could be improved by increasing the temperature to reflux but at the expense of increased weight losses.

The initial and final activities on the hulls are given in Table 16.

If used on a large scale the cerium(IV) would be produced electrolytically from cerium(III) and some regeneration of the spent reagent would be possible and economically desirable. The waste management of the effluents has not been studied in detail but could involve reclamation of the nitric acid by, for example, evaporation.

6.2.3 Alkaline Permanganate (A.P.)

This reagent has been widely used for the decontamination of stainless steels[16-20]. It is normally used as the first stage of a two-stage process, the permanganate oxidising the chromium in the surface layers of the corroded steel to the hexavalent state, which is removed by the alkali as dichromate. This action modifies the residual corrosion product layers on the steel, rendering them more soluble in the second stage for which a variety of solutions have been proposed, mostly based on ammonium citrate together with some other complexing agent. The second stage solution used in the present work was based on the CITROX formula and consisted of a mixture of diammonium hydrogen citrate (5%) and oxalic acid (2.5%). It was designed to neutralise residual alkalinity, to dissolve manganese dioxide from the AP stage, to complex the dissolved metals, and to retard redeposition. The permanganate solution consisted of 3% potassium permanganate in 10% sodium hydroxide; the hulls were given one-hour contacts at reflux. The treatment with the CITROX-type solution involved one-hour contacts at 80-90°C. Weight losses from the hulls were very low.

The results are given in Tables 17 and 18 which show that for 5.6% and 7.3% burn-up hulls, high levels of removal of both α-emitters and fission products (including Ru-106) were achieved. Fission product removals from 4.1% burn-up hulls were generally poor although earlier work with other hulls from the same batch gave results more in line with those of the higher burn-up batches.

Reducing the A.P. concentrations to a half to cut down the salt content of the effluents had a deleterious effect on the decontamination.

Table 19 shows the performance of the individual stages of the process for two high burn-up hulls and shows the CITROX stage to be more important for α-emitters than for fission product removal.

The process is highly effective for decontaminating hulls but suffers the disadvantage that large volumes of secondary waste, including some of high salt content requiring special effluent treatment, could be generated.

6.2.4 Oxalic Acid

Oxalic acid is another reagent that has found extensive use in steel decontamination. It has frequently been used in conjunction with

other substances, including ammonium citrate, hydrogen peroxide, ammonium fluoride and gluconic acid[3,19,21-24].

Used on its own for hull decontamination it was found to be rather corrosive, and the addition of hydrogen peroxide as a corrosion inhibitor gave improved activity removal as well as lower weight losses.

The mixture used for most of the work was 0.5M oxalic acid, 1M hydrogen peroxide. Contactings (up to three in number) were carried out at reflux, their duration varying from 2 to 6 hours. Reducing the acid concentration to 0.1M produced only a small effect. In the presence of hydrogen peroxide most of the activity removal occurred during the first contacting whereas the oxalic acid alone the relative proportion of activity removed during the second contacting was somewhat higher. This was especially so for Ru and Cs activities.

The results for oxalic acid decontamination are given in Tables 20 and 21.

6.2.5 Other Reagents

6.2.5.1 LOMI reagents

The use of low oxidation state metal ions has featured much in recent literature[25,26,27] as the basis of decontaminants for the primary coolant circuit of Boiling Water Reactors and of the Steam Generating Heavy Water Reactor at Winfrith. These "LOMI" reagents operate by reducing the ferric component of the oxide cruds to the ferrous state which goes into solution and is complexed by a chelating agent. The most commonly used LOMI reagent consists of a mixture of vanadous formate, formic acid and picolinic acid, typical concentrations being: V^{2+} 4 x 10^{-3}M, HCOOH 4 x 10^{-2}M, and picolinic acid 2.5 x 10^{-2}M. Because of the low reagent concentrations used, the decontamination liquors can be readily cleaned up by an ion-exchange process.

To examine the feasibility of applying these reagents to hull decontamination, some vanadous formate was prepared electrolytically by the method of Bradbury et al[28] and mixed with picolinic acid in the appropriate proportions. Hulls were treated with the solution for periods of up to six hours at 80-85°C and at reflux. The results given in Table 22 show that the LOMI reagent only removed 80-90% of the α-emitters and less than 20% of the fission products, irrespective of

the duration of the contact, when used at concentrations similar to those employed for cleaning water reactor coolant circuits. The weight losses were about 1%. At reflux temperature the weight losses were surprisingly much lower, although the activity removals, apart from Cs-137, were not significantly altered.

When the LOMI reagent concentration was increased by a factor of 5, the removals of α-emitters were greatly reduced and the separation of a reddish-brown solid from the solution was observed.

6.2.5.2 Nitric Acid-Permanganate (The POD Process)

This process[27] was developed in conjunction with the application of LOMI reagents to PWR circuits where an oxidative pretreatment was necessary to deal with high chromium-content oxides. It consisted of a three-stage process: (i) $KMnO_4$ (0.006M)/HNO_3 (0.005M) for 5-24 hours reflux, (ii) oxalic acid (0.0156M)/HNO_3 (0.018M) for 1-2 hour reflux, and (iii) oxalic acid (0.0025M)/citric acid (0.0025M) for up to 6 hours reflux. When applied to hulls the POD process on its own gave poor activity removals for all isotopes except those of caesium. In one case the decontaminated hull was treated with LOMI reagent, but only for ruthenium was the additional activity removal significant.

6.2.5.3 Formic Acid-Formaldehyde

A mixture of formic-acid and formaldehyde has been described[29] as suitable for decontaminating steel surfaces such as those in reactor cooling circuits. The mixture gave very poor results when applied to fast reactor hulls, the α-emitter removals being around 20% and the fission product removals much lower.

6.2.6 Comparison of Principal Decontamination Routes

Fig. 15 compares the performance of the principal decontamination routes in terms of α-activity removals. For each route the range of typical values of percentage removals, residual activities and weight losses are plotted against burn-up.

The percentage removals show a wide spread of values, whereas the residual activities on the hulls generally lie in a narrow band corresponding to 0.1 to 0.3 $\mu Ci/g$, irrespective of burn-up or decontamination route. The main departure from this behaviour is the 4.1% burn-up hulls treated with nitric acid/cerium (IV) which had much higher residual activities. The samples involved all originated from a single hull which had an abnormally high Mn:Co ratio. The results cannot be considered typical ones since earlier work with several different hulls from the same batch had given values more in line with the general trend. Lowest weight losses were recorded with the alkaline permanganate and oxalic acid-hydrogen peroxide reagents.

6.2.7 Decontamination of Complete Hulls

A short programme of work on the decontamination of complete hulls by nitric acid was undertaken. The objective was to confirm that the findings obtained using small slices of hull were applicable to whole hulls.

Four batch E hulls were each contacted for 3 two-hour periods at reflux with 9M nitric acid. The experimental conditions followed closely those described in Section 5.2 but additionally, each of the solutions generated was passed through a filter unit consisting of a 0.4 µm HAWP Millipore filter backed by a 0.7 µm glass fibre paper to remove any fine particulate material the solutions might contain.
Three of the hulls used were 50 mm long; the other was 25 mm (see Table 23). The weight losses from the hulls at each stage of the decontamination could be measured much more accurately because of the greater size of the samples.

Hull E/3A/4 was photographed at each stage of the decontamination to monitor any visual changes occurring during the process. The photographs showed that the initially glossy surface of the hull became progressively more matt and pitted (see Fig.16). No other alteration of the surface was apparent. Sample E/3A/5, on the other hand, initially appeared shiny with large areas of black deposit. After the first nitric acid contacting, most of this black material had gone. The few grey patches that remained were removed by the second nitric acid treatment leaving the hull clean and shiny.

The flasks employed in the decontaminations were degreased prior to use by treatment with Genklene and with concentrated nitric acid. In spite of this, high levels of activity were found to have deposited on the walls during some of the contactings. The activity included some fine particulate materials. It was found that treatment of the flasks with a mixture of nitric and hydrochloric acids containing either hydrofluoric acid, or a carrier solution (a mixture of inactive compounds of the principal fission product elements) reduced the deposited activities to lower levels but did not always bring about complete dissolution of the particulates. This could normally be achieved by an alkali treatment involving reflux with 2M potassium hydroxide/5% potassium periodate.

The weight losses from the hulls during decontamination were, with one exception, in the range 1.2-2.7%. This compares with values of 0.7-3.5% obtained from 1 mm slices of hulls from the same batch (see Tables 13 and 24). The exception was E/2A/2 which had a much higher weight loss(6.0%); this was due to a considerable quantity of dense black particulate material liberated during the first contacting. Treatment of this with the nitric acid/hydrochloric acid/mixed fission product carrier solution extracted large quantities of ruthenium and caused the material to become less granular. For complete dissolution the alkali treatment was required, and the resulting solution contained further large quantities of ruthenium. As there had been no observable change in the appearance of the outer surfaces of the hull during the nitric acid contacting, it is thought that the black ruthenium-rich material may have originated from the inner surfaces of the hull. The nature of this material suggested it might have been a dissolver insoluble type of residue. Because it had not been compacted in a centrifuge, it would not have been subjected to a sufficiently high heat concentration to bring about sintering, and consequently would not have reached the degree of insolubility often associated with this type of material.

In the small-scale work it had been thought that some of the weight loss could be attributed to the attack of acid on the freshly cut, bare metal ends. Whereas this may have been the case with nitric acid in the presence of ceric ions, it now seems less likely with nitric acid on its own since the weight losses from whole hulls with no untreated bare ends were the same as those from the small slices.

The quantities of activity deposited on the flasks and in the particulate materials are included in the analytical figures. Table 24 describes the overall decontamination performance in terms of percentage removals and activities before and after decontamination. Comparing the percentage removals from whole hulls with those obtained from 1 mm slices of hulls from the same batch (Table 13), a considerable improvement for the fission products was observed whereas the removal of α-emitters was slightly lower. In terms of residual activity the α-emitters were a factor of 3-4 times higher on the whole hulls. Table 25 describes the percentage removals at each stage of the decontaminations. The bulk of the α-activity removal (75-95%) occurred during the first contacting: the second contacting accounted for most of

the rest of the activity removed and the third contacting was
essentially a polishing stage. Fission product removals were more
variable and tended to be spread out more evenly over the three
contactings.

The total activities (in μCi/g at completion of irradiation) of the
principal isotopes found on the hulls all lay (except for those of
E/2A/2) within the same ranges as those for the 1 mm and 10 mm slices -
see Fig.11. This further vindicates the use of small slices, as
discussed in section 6.1.4.

The iron contents of the solutions from the decontaminations were
determined by a colorimetric method based on o-phenanthroline.
Preliminary results indicated that only about a third of the weight loss
from the hulls could be attributed to dissolved steel. Similar
measurements on solutions from the decontamination of 1mm slices from
the same batch of hulls gave similar figures.

Examination of the filters through which the solutions were passed
indicated the presence of small amounts of particulates. γ-Interrogation
of these showed Ru-106 to be the main radionuclide present, together
with smaller quantities of Sb-125, Cs-137 and Cs-134 in order of
decreasing activity.

The presence of particulates was sought in some of the solutions
arising from the decontamination of 1mm hull slices. Visually the
solutions appeared quite clear and particle free, but when passed
through a 0.65 μm filter, up to 20 small, visible, black particles were
collected on the filter for each ~ 50 mL solution examined. These were
γ-counted and found to contain principally Ru, Sb and Cs, together with
small amounts of Mn-54 and Co-60. The ratios Sb-125:Ru-106:Cs-137 for
these particles were found to be very similar to those for typical
dissolver insoluble particulates[30] (see Table 26). It has been shown,
inactively[30], that simulant dissolver insoluble particulates suspended
in solution in contact with stainless steel enhances the corrosion of
the steel, especially at grain boundaries. In the light of these
observations, it seems probable that the particles collected on the
filters were dissolver insolubles, the Mn and Co originating from the
corrosion of the activated cladding.

The problem of the plate out of activity on the flasks, although
observed in the very early work on 1 mm slices, was much less
significant in later work.

The preliminary work on the treatment of whole hulls with 9M nitric
acid at reflux has shown that the levels of decontamination are of the
same order of magnitude as those obtained with 1 mm hull slices; that
the weight losses incurred are also very similar, and that part of the
weight loss can occur through the removal of dissolver insoluble-type
material. The specific activity levels on the hulls are in the same
ranges as those found on both 1 mm and 10 mm slices.

6.3 Immobilisation

The inactive tests showed that about 40g of hulls could be incorporated
in the 50 mm cylindrical products, whilst still retaining adequate strength
for handling in-cell. When higher weight loadings were used there was
insufficient cement between the hulls and the product was very brittle. It
was also found that the vibrator had to be tuned to minimise voids in the
product (Fig.17) and to get a fairly uniform distribution of hulls in the
sample.

Generally it has been possible to produce substantially void-free
products but changes in the composition of the grout have a significant
influence on the structure of the product. Thus with inactive sections of
PFR hulls immobilised in BFS/OPC the distribution of hulls is greatly
influenced by the frequency of vibration. In Figure 18 comparison is made
between hulls in sand/OPC and BFS/OPC vibrated at the same frequency. In the
sand/OPC the hulls are distributed in such a way that they are reasonably
uniformly dispersed but with BFS/OPC at the same frequency, the hulls are
very unevenly distributed with a high loading at the bottom of the sample.

Visual inspection of the surfaces of the immobilised waste forms showed
that they were free of cracks, but some of the sand/OPC samples had small
holes in the sides (Table 2).

6.4 Leach Testing

Radiochemical analysis of the hulls prior to immobilisation showed that
the predominant nuclide activities available for leaching were the fission
products Sr-90, Ru-106, Sb-125, Cs-134 and Cs-137 and the actinides. Other
nuclides would also be available, but they were not readily detectable on a
routine basis. The activation products Mn-54 and Co-60 were not regarded as
being readily available for leaching, unless appreciable corrosion of the
hulls occurred.

The results are expressed as plots of the cumulative fraction of the
activity released (Figs. 19-22), and of the incremental leaching rate (see
Fig. 23), both as a function of the time of leaching. The cumulative
fraction (F) is defined as,

$$F = \frac{\sum a_n}{A_o} \times \frac{V}{S} \ cm$$

and the incremental leach rate (R_n) as,

$$R_n = \frac{a_n/A_o}{(S/V)t_n} \ cm.d^{-1}$$

where a_n = leachate activity

A_o = incorporated activity

S = exposed surface area of specimen (cm^2)

V = specimen volume (cm^3)

n = number of leachant renewals

t_n = duration of nth leachant renewal period (d)

The cumulative fraction of Sr-90 activity leached is given in Fig. 19,
whilst Figs. 20, 21 and 22 are the corresponding curves for Cs-137, Ru-106
and Sb-125 respectively.

Analysis of the leachates from the immobilised samples showed that the
only significant activities leached were Sr-90 and Cs-137. (Cs-134 is also
leached but since it behaves in the same manner as Cs-137 only data for the
latter are reported). Ru-106 and Sb-125 are also leached but only from the
samples made using sand/OPC.

In order to establish the contribution being made by the matrix material
in retaining the available nuclide activities, comparison was made with data
obtained from leaching bare material. This material was leached under
similar conditions to those of the immobilised samples with the leachants
being either distilled water or water previously equilibrated with the
appropriate cement grout.

The results from leaching the bare hulls (Figures 19-22) showed there
was little difference in leaching of the respective nuclides between the
three leachants that had been used. However, it would appear that Sr-90 and
Cs-137 were more readily leached than Ru-106 and Sb-125. This may be
attributable to both the latter being predominantly present in an insoluble
form such as the dissolver residue which comprises the noble metal fission
product alloys Mo, Tc, Ru, Rh and Pd.

Comparison of the immobilised products shows there was very little
difference in the leaching behaviour of Cs-137 between sand/OPC and BFS/OPC
(Fig. 20). After 100 days of the ISO test the cumulative fraction of Cs-137
leached from the immobilised products averaged 1-2%. For the bare material
the fraction leached was about an order of magnitude greater, showing the
difference that could be attributed to the effect of the matrix. However, it
is worth noting from the leach rate curves (Fig. 23) that the biggest
differences between the bare and the immobilised products occurred in the
early stages of leaching. As the leach tests progressed, so the leach rates
of Cs-137 from both the bare and the immobilised samples tended towards a
similar value (compare the rates at 100 days).

The results for Sr-90 (Fig. 19) imply that BFS/OPC provides a better
retention for this nuclide than sand/OPC by about an order of magnitude.
After 100 days of the ISO test the cumulative fractions leached averaged
about 10%, 1% and 0.1% for the bare material, sand/OPC and BFS/OPC,
respectively. Again the greatest differences in the results occurred in the
early stages of the test where leach rates for the bare material were
significantly higher.

The reason for the leaching of Ru-106 and Sb-125 from sand/OPC and not
from BFS/OPC is not obvious (Figures 21, 22). However the cumulative
fraction leached from the bare hulls tended to be lower by about an order of
magnitude than for Sr-90 and Cs-137, suggesting that they all do not arise
from the same source. Sr-90 and Cs-137 are likely to arise, at least in
part, from dissolver solution which has dried on the hulls following fuel
dissolution, whereas the Ru-106 and Sb-125 are major components in the
dissolver residue which may adhere to the hulls.

Apart from the first one-day leach, it was found that the individual
nuclide activities appearing in sequential leachates were similar,
irrespective of the leaching period. Because of this similiarity in the
activity levels, it was assumed that a 'steady state' release was fairly
quickly established, and in order to understand the appertaining mechanisms,
future studies will need to include such parameters as sorption and
solubility.

The level of α-activity leached from the immobilised waste forms during the period of the test was below the limit of detection ($\sim 5 \times 10^{-10}$Ci) and the amount of activity leached from the bare hulls indicated that only about 8% of the total α-activity associated with the samples was available for leaching.

7. CONCLUSIONS

CHARACTERISATION

1. The level of α-activity on the hulls varied from 0.004-0.2 Ci/Kg, the principal constituents being Pu-239, Pu-240, Pu-238, Am-241, Cm-242 and Cm-244.

2. The predominant fission product was Ru-106 (1-400 Ci/Kg) followed by Sb-125 (0.4-5 Ci/Kg) and Cs-137 (0.02-0.5 Ci/Kg). (The figures refer to the time of completion of irradiation). Cs-134, Ce-144 and Tc-99 were also present as well as traces of Eu-155, Eu-154 and Ag-110m.

3. The main activation products were Mn-54 (up to 250 Ci/Kg) and Co-60 (generally in the range 1-5 Ci/Kg). Co-58 was also detected in some high burn-up hulls. Levels of up to 0.005 Ci/Kg of C-14 were found in a range of samples.

4. Profiles of the principal activity levels along lengths of about 17 hulls have been prepared. Substantial variations of, for example, ruthenium along a hull may result from the presence of particulates deposited from the dissolver liquors.

5. Levels of I-129 and Np-237 were calculated to be too low for ready measurement.

6. Actinides were confined to the top 2-3 microns of the hull surface and the fissile material generally corresponded to < 0.1% of the initial fuel.

7. Evidence from total fissile measurements, fission track autoradiography, Pu/Am ratios, and the measurement of the activities on unirradiated and pin-end hulls suggested that deposition of activity from the dissolver liquors accounted for significant proportions of the activity on the hulls.

DECONTAMINATION

8. Small scale experiments showed that the level of α-activity on the hulls could be reduced to below 0.3 Ci/tonne by a number of different reagents. This was independent of the reagent type or the burn-up (up to 7.3%) of the fuel the hulls had contained.

9. The preferred reagent was 9M nitric acid at reflux. This should produce a low-salt effluent which can be recycled to the reprocessing plant.

10. The results of the decontamination of complete hulls by refluxing with nitric acid generally supported the findings from the smaller scale experiments. The weight losses were very similar; the fission product removals were higher and more consistent, but the α-removals were slightly lower. There was evidence that some of the weight losses arose through the removal of particles of dissolver insoluble materials. Further work with whole hulls is needed to provide a more complete understanding of the decontamination process.

11. The use of a decontamination step could permit shallower burial and hence a cheaper disposal route for the hulls.

IMMOBILISATION

12. The cumulative fraction of Cs-137 leached from the immobilised samples after 100 days of the ISO test averaged about 1-2% and there was no significant difference between sand/OPC and BFS/OPC as the matrix material.

13. The cumulative fraction of Sr-90 leached from the immobilised samples after 100 days of the ISO test averaged about 1% for the sand/OPC and about 0.1% for the BFS/OPC matrix. Obviously the latter is better for retaining Sr-90, but the reason needs further elucidation.

14. The cumulative fraction of Cs-137 and Sr-90 leached from the bare material after 100 days averaged about 10-20%. This implied that the remaining activity was not readily available for leaching during the period of the test and consequently the effectiveness of the matrices could be overestimated.

15. Comparison of the leach rates of Cs-137 for the bare and immobilised waste showed that the biggest difference occurred in the first few days of leaching. As the leach testing progressed the leach rates tended to become similar.

16. Ru-106 and Sb-125 can be leached from sand/OPC but not from BFS/OPC matrices.

17. Only about 8% of the α-activity was available for leaching. The actual amounts leached from the cemented waste forms were below the limits of detection.

18. From visual observations and leach testing studies cement appears to be a suitable medium for immobilisation of hulls, the BFS/OP matrix being somewhat better than the sand/OPC.

19. Further evaluation of the waste forms needs to be carried out. This should include: autoradiography, equilibrium leach testing, degree of sorption on the matrix, and the activity release on heating.

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Table 1 Details of Hulls and Composition of the
Fuel they had Contained

Batch Number	Approximate Number of Hulls	Length of Hulls (mm)	Burn-up of Fuel (%)	% Pu in Dissolver Solution	Date of Removal from Reactor
A	50	50	5.6	14.1	9.3.82
B	35	25-150*	3.9-5.6	14.1-18.5	6.9.79)+ 9.3.82)
C	40	50-75	7.3	18.2	6.9.81
D	50-60	50	0	19.0	Not irradiated
E	250	25++	4.1	19.2	20.2.78

Notes: * The majority of the hulls in this batch were 50-75 mm long
+ Mixed batch of hulls originating from 2 sub-assemblies
++ Contained also some fuel pin end pieces 90-110 mm long.

Batch Number	Burn-up (%)	Activity of Fuel in Curies after 5 Years' Cooling					
		Pu-238	Pu-239	Pu-240	Am-241	Cm-244	Total α
E	4.1	289	641	642	533	2.3	2773
A	5.6	326	455	516	493	14.3	1826
C	7.3	484	584	679	751	20.8	2553

Table 2   Samples Prepared for Leach Testing

Sample No.	Hull Material	Matrix	Length of Cure Before Leach Testing	Visual Inspection
		Immobilised Hulls		
HLJ	92.5g batch A	BFS:OPC:water	91 d	No cracks, 3 very small holes in side
HLK	35.83g batches A,B,C	BFS:OPC:water	90 d	No cracks, top edge chipping
HLL	33.61g batches A,B	Sand:OPC:water	41 d	
HLM	37.19g batches A,B + 47.33g blank stainless steel	BFS:OPC:water	118 d	No cracks
HLN	93.62g batch C	Sand:OPC:water	41 d	No cracks, many hole in side, side and to edge crumbling
HLU	99.80g batch E/1	Sand:OPC:water	28 d	No cracks, hole in side, hulls visible
HLV	99.70g batch E/1	BFS:OPC:water	91 d	No cracks
HLW	100.26g batch E/2	Sand:OPC:water	28 d	No cracks, some hole in side, hulls visib
HLX	99.88g batch E/2	BFS:OPC:water	91 d	No cracks
HLY	100.26 batch E/3	Sand:OPC:water	28 d	Four blow holes in sides, no cracks
HLZ	100.06g batch E/3	BFS:OPC:water	91 d	No cracks
		Bare Hulls		
HUL5	5.72g batch A	NIL	-	
HUL6	5.65g batch C	NIL	-	
HUL7	7.26g batch E/1	NIL	-	
HUL8	7.24g batch E/2	NIL	-	
HUL9	7.21g batch E/3	NIL	-	

Table 3   Total Fissile Content of Fast Reactor Hulls

Hull Identification No/Ring	Remarks	Burn-Up (%)	Approximate Mn-54:Co-60	Radiation at 15cm rad/h βγ	Y	Fissile Content as Pu-239 mg/Kg	Ci/Kg x10³	Pu as % of Original Fuel
C2/B		7.3	74	1.4	0.28	90	5.6	0.012
C3/A		7.3	8	>5	1.8	390	24	0.06
C3/J		7.3	7	3.7	0.8	7·)	14	0.03
A1/A		5.6	6	4.5	1.1	340	21	0.05
A1/J		5.6	5	0.8	0.19	50	3.1	0.008
A3/J		5.6	11	0.22	0.05	20	1.3	0.004
A5/G	Spacer	5.6	~0	>5	2.6	810	51	0.14
E1/2/F	Shiny	4.1	3	0.025	0.008	15	0.9	0.002
E2/3/B	Black	4.1	108	0.7	0.1	1365	85	0.23
D1/B	Unirradiated	0	-	1.2	0.25	520	33	0.07
D1/H	Unirradiated	0	-	0.22	0.05	125	8	0.02

Table 4   Percentage Activity Composition of α-Emitters as Determined by α-Spectrometry

Well No	Burn-Up (%)	ORGANIC PHASE			AQUEOUS PHASE						OVERALL						Date of Count
		Pu239+240	Pu238	OTHER	Pu239+240	Pu238	Am241	Cm243+244	Cm242	OTHER	Pu239+240	Pu238	Am241	Cm243+244	Cm242	OTHER	
B1/A	0	81.0	19.0	-	18.0	4.2	72.8	-	-	5.0	45.9	10.8	40.8	-	-	2.1	27.6.84
B2/3/B	4.1	78.6	21.4	-	8.8	2.4	85.8	1.3	1.7	-	56.5	15.4	27.2	0.4	0.5	-	12.9.84
B1/10/A	4.1	79.0	21.0	-	16.0	4.3	75.7	-	-	4.0	55.7	16.8	28.0	-	-	1.5	27.6.84
B1/12/A	4.1	80.0	19.0	1.0	14.0	3.3	77.7	-	-	5.0	56.7	13.0	30.4	-	-	1.9	27.6.84
B1/12/F	4.1	87.6	12.4	-	39.7	5.6	54.0	0.3	0.4	-	80.6	11.4	7.9	0.04	0.06	-	12.9.84
A1/B	5.6	72.0	28.0	-	3.7	1.4	51.2	0.3	42.9	0.5	38.8	15.1	24.9	0.15	20.9	0.15	16.2.84
A3/B	5.6	72.1	27.9	-	20.2	7.8	47.5	2.6	20.6	1.3	52.2	20.5	18.0	1.0	7.8	0.5	12.9.84
A5/A	5.6	72.2	27.8	-	-	-	59.5	1.4	40.5	-	40.9	15.7	25.8	-	17.6	-	16.2.84
C2/F	7.3	72.6	27.6	-	9.7	3.7	51.3	-	35.3	-	40.9	15.6	25.8	-	17.7	-	16.2.84
C3/B	7.3	74.0	26.0	-	16.8	5.6	52.4	-	-	25.0	53.2	18.7	20.6	-	-	7.5	27.6.84
C6/F	7.3	77.2	22.7	0.1	16.5	4.8	67.6	1.4	9.7	0.2	58.1	17.1	21.3	0.4	3.0	0.1	12.9.84

- 37 -

Table 5 Comparison of Measured α-Constituents with FISPIN Predictions at 16.2.84

| Hull No | % Composition by Activity | | | | | | Ratio |
	Pu239+Pu240	Pu238	Am241	Cm243+Cm244	Cm242	OTHER	Am241: Pu238: (Pu239+Pu240)
A1/H	38.8	15.1	24.9	0.15	20.9	0.15	1 : 0.6 : 1.6
A3/B	47.0	18.5	16.2	0.8	17.1	0.4	1 : 1.1 : 2.9
A5/A	40.9	15.7	25.8	-	17.6	-	1 : 0.6 : 1.6
FISPIN BATCH A	46.7	16.0	18.7	0.8	17.8	-	1 : 0.9 : 2.5
C2/F	40.9	15.6	25.8	-	17.7	-	1 : 0.6 : 1.6
C3/H	50.3	17.7	19.5	-	12.5	-	1 : 0.9 : 2.6
C4/F	55.6	16.4	20.4	0.4	7.1	0.1	1 : 0.8 : 2.7
FISPIN BATCH C	52.3	19.6	22.8	1.9	4.3	-	1 : 0.9 : 2.3
E2/3/D	56.0	15.3	27.0	0.4	1.3	-	1 : 0.6 : 2.1
E1/10/A	55.7	14.8	28.0	-	-	1.5	1 : 0.5 : 2.0
E1/12/A	54.3	12.9	30.1	-	-	1.9	1 : 0.4 : 1.8
FISPIN BATCH E	59.5	15.3	25.1	-	-	-	1 : 0.6 : 2.4

Table 6   Isotopic Composition of the Plutonium on Fast Reactor Hulls

Hull Identification Number/Ring	Burn-up (%)	Atom Percent				
		Pu-238	Pu-239	Pu-240	Pu-241	Pu-242
C2/F	7.3	0.21	74.65	22.29	2.21	0.66
A1/H	5.6	0.25	73.38	23.32	2.37	0.68
A5/A	5.6	0.23	73.69	23.09	2.33	0.67
E1/14 (black)	4.1	0.23	75.64	21.48	2.08	0.57
E1/15 (black)	4.1	0.17	75.53	21.62	2.11	0.58
E1/16 (shiny)	4.1	0.19	75.80	21.36	2.09	0.57

Table 7   γ-Emitting Activation Products

Hull No.	Burn-up (%)	Mean Activities (µCi/g) at Time of Completion of Irradiation			Mn-54:Co-60 Across Hull
		Mn-54	Co-60	Co-58	
C1	7.3	7,370	4,940	198,863	1 45 → 1.53
C2	7.3	215,943	2,889	3,308,815	73.9 → 76.3
C6	7.3	126,385	1,670	1,958,297	74.0 → 78.0
A1	5.6	21,911	4,107	88,988	4.7 → 5.9
Ring*					
A5 (i)	5.6	N.D.+	2,548	N.D.	~0
(ii)		1,249	13,716	N.D.	0.08
(iii)		1,495	11,817	N.D.	0.13
(iv)		1,613	3,380	N.D.	0.48
(v)		1,827	3,365	N.D.	0.54

+N.D. - not detected.

*Location of rings along hull A5 (see also Fig. 7):

    (i) Below spacer
   (ii) Around crimp
  (iii) Around location of Mo wire disc
   (iv) Just above spacer
    (v) Above spacer, at other end of hull.

Table 8 Analysis of Hulls Used for C-14 Determination

Hull	Mn-54:Co-60	ACTIVITIES μCi/g								
		Gross α	Ce-144	Sb-125	Ru-106	Cs-137	Cs-134	Mn-54	Co-60	C-14
C7/A	1.3	13.8	-	2,657	143,779	34	-	7,077	5,6:2	0.15
C4/B	17.7	22.2	564	825	12,589	272	37	27,485	1,553	4.7
C4/C	17.9	17.4	582	557	10,714	220	23	20.837	1,167	3.2
C4/J	17.4	15.7	349	612	11,036	220	27	22,848	1,314	0.3:
C6/P	75.7	2.9	-	1,834	15,937	220	36	152,650	2,016	0.86
C6/L	76.0	6.4	-	2,057	18,517	143	-	182,080	2,397	0.82
C6/N	76.8	12.8	-	1,052	14,060	128	-	84,188	1,096	1.0
E2/2/D	9.5	23.8	-	2,622	347,197	112	-	49,729	5,262	0.06
E2/2/B	9.9	6.3	-	738	23,214	36	-	32,931	3,329	0.09
E3/3/E	40.2	5.9	-	138	22,330	342	-	59,067	1,468	0.31
E3/3/C	42.0	12.8	-	313	56,477	361	-	63,764	1,518	0.32
E3/3/B	43.7	48.0	-	1,456	294,543	501	-	79,689	1,825	0.17
E3/3/A	44.9	141.8	-	3,993	898,085	571	-	66,137	1,473	0.40
E3/4/E	91.9	6.4	951	306	10,849	302	56	108,113	1,177	0.44

TABLE 9

Summary of the Mean Activities on PFR Hulls

The figures represent averages of the measurements on the individual rings of each hull

Hull No.	Burn-up (%)	Gross g	Sb-125	Ru-106	Cs-137	Cs-134	Ce-144	Eu-155	Sr-90	Mn-54	Co-60

NM — Not measured

Table 10 Activities Associated with End Piece Bulls

Bull No./ Ring	Distance from closed end mm	Activities µCi/g							Mn:Co
		Gross α	Sb-125	Ru-106	Cs-137	Sr-90	Mn-54	Co-60	
E1/10/A	21	88	674	48409	217	753	10869	3143	3.46
E1/10/E	27	272	839	72627	909	NM	11206	3030	3.70
E1/10/J	93.5	77	2471	299070	157	NM	21808	2365	9.22
E1/10/F	99.5	52	1376	133744	122	44	23632	2347	10.07
E1/13D/A	28	15	190	8264	301	28	14279	4592	3.11
E2/4/D	4	6	690	24808	25	5	10243	3604	2.84
E2/4/F	7	7	648	39188	41	NM	11322	3620	3.13
E2/4/A	30	84	4009	348588	163	35	13878	3164	4.39

NM - Not Measured".

Total Fissile Content of End Pieces

Bull No./ Ring	Distance of Ring from Closed End mm	Fissile Content as Pu-239 µg/g
E1/10/B	22.5	1270
E1/10/G	98	360

Table 11 **Comparison of the Mean Activity Levels of the Batches of Hulls**

Batch	Burn-up (%)	Mean Activities (µCi/g on 17.5.84)			
		Gross α	Sb-125	Ru-106	Cs-137
D	Unirradiated	19.3	316	2972	78
A	5.6	9.75	604	4661	392
C	7.3	7.95	1676	7734	430

Ratios of Activities of Fission Products

	Sb-125	Ru-106	Cs-137
Batch C dissolver solution	1	12.2	13.6
Batch A dissolver solution	1	18.3	10.7
Batch D on hulls	1	7.9	0.3
Batch A on hulls	1	7.7	0.6
Batch C on hulls	1	4.6	0.2

Table 12   Details of Typical Nitric Acid Decontaminations for Hulls of Two Different Burn-ups

% Activity Removals

	Gross α	____	Fission Products					Activation Produc		
		Sb-125	Ru-106	Cs-137	Cs-134	Ce-144	Ag-110m	Mn-54	Co-60	Co-5
Hull No./Ring:A1/F5.6% Burn-up					9M HNO$_3$	3 x 6 hr Contactings at Reflux				
1st Contacting	91.8	76.5	52.7	35.8	–	22.7	100	1.4	1.8	0
2nc Contacting	3.8	4.0	10.6	5.2	–	3.6	0	0.6	0.7	0
3rd Contacting	2.5	3.3	9.0	6.0	–	4.5	0	0.8	0.8	0.8
Total Removal	98.1	83.8	72.3	47.0	–	30.8	100	2 8	3.3	0.8
Hull No./Ring: C2/H 7.3% Burn-up					9M HNO$_3$	3 x 6 hr Contactings at Reflux				
1st Contacting	9J.6	78.0	35.7	49.3	51.9	100	–	5.8	7.3	6.9
2nd Contacting	2.8	19.4	12.9	46.2	48.1	0	–	3.0	2.8	2.8
3rd Contacting	0.1	2.6	5.1	4.5	0	0	–	2.9	2.9	2.9
Total Removal	93.5	100	53.7	100	100	100	–	12.7	13.0	12.7

Table 13   Decontamination of PFR Hulls with Nitric Acid

Hull Identi- fication	Burn-up (%)	Mn-54:Co-60	PERCENTAGE REMOVALS							Weight Loss (%)	Calculated Thickness Removed (um)
			Gross a	Sb-125	Ru-106	Co-137	Ce-134	Ce-144	Sr-90		
9M HNO₃ - 3 x 6 hour contact at reflux											
C2/E	7.3	74.7	98.6	100	73.6	100	100	100	NM	21.0	23
C2/B	7.3	73.7	93.5	100	53.7	100	100	100	NM	13.1	15
C2/I	7.3	75.9	96.3	100	58.5	100	100	-	NM	13.5	14
C2/J	7.3	74.1	98.9	100	69.4	100	100	100	NM	12.4	14
A1/B	5.6	6.0	97.4	86.2	79.5	67.1	-	100	NM	3.5	4
A1/C	5.6	5.6	93.1	100	73.5	44.0	-	100	NM	3.2	3
A1/E	5.6	5.5	98.9	100	80.4	64.6	-	100	NM	4.3	5
A1/F	5.6	5.3	98.1	83.8	72.3	47.0	-	30.8	NM	2.7	3
A3/E	5.6	10.7	97.5	100	88.1	46.0	100	1	NM	2.7	3
A5/L	5.6	0.08	98.7	100	88.2	100	-	100	NM	3.1	*
A5/Q	5.6	0.13	98.9	100	94.2	100	-	-	NM	10.7	*
9M HNO₃ - 3 x 6 hour contacts at 60°C											
A3/A	5.6	12.0	81.3	61.8	11.1	23.0	100	100	NM	0.4	<0.5
A3/B	5.6	9.8	83.9	63.5	15.1	14.9	100	100	NM	0.2	<0.2
9M HNO₃ - 3 x 2 hour contact at reflux											
C6/A	7.3	74.1	98.6	74.7	57.0	51.9	100	100	78.7	3.0	3
C6/C	7.3	74.7	94.7	87.7	46.8	61.0	100	100	NM	3.0	3
A4/E	5.6	11.3	91.6	100	84.4	93.6	100	100	NM	7.0	9
A4/G	5.6	11.2	96.6	100	94.4	96.5	100	100	84.4	8.0	9
E1/1/C	4.1	68.1	99.71	91.1	92.0	50.6	37.8	35.2	NM	1.6	2
E1/1/F	4.1	70.2	99.88	96.0	97.1	58.8	56.1	77.7	51.5	3.5	5
E1/2/A	4.1	2.1	91.0	100	70.1	11.4	100	-	NM	1.0	1
E1/2/C	4.1	2.5	98.2	100	34.6	12.3	100	100	NM	0.7	1
E1/10/D	4.1	3.8	99.67	69.7	62.1	96.3	100	100	NM	1.4	*
B1/J	0	-	94.9	89.4	71.3	95.3	100	90.5	NM	1.2	1
9M HNO₃ - 3 x 1 hour contact at reflux											
C4/A	7.3	17.9	98.9	100	69.9	56.9	100	100	NM	1.0	1
A3/F	5.6	10.4	91.2	100	46.7	27.2	100	100	NM	1.4	2
E2/3/A	4.1	108.5	96.3	90.4	85.8	88.3	100	100	92.1	12.9	18
E2/3/E	4.1	112.0	98.8	90.4	52.2	100	100	100	NM	16.2	23
6M HNO₃ - 3 x 6 hour contact at reflux											
C6/E	7.3	76.9	97.1	83.1	59.1	37.6	100	100	NM	4.9	6
C6/G	7.3	76.4	97.2	100	59.0	46.8	100	100	97.9	4.0	5
6M HNO₃ - 3 x 2 hour contact at reflux											
C3/C	7.3	8.0	93.8	86.2	63.1	40.0	-	57.8	NM	2.8	3

* Not calculated, different geometry of ring.
NM Not measured.

Table 14   Activities on Hulls Before and After Decontamination with Nitric Acid

Hull Identi-fication	ACTIVITY BEFORE DECONTAMINATION (µCi/g HULL)							ACTIVITY AFTER DECONTAMINATION (µCi/g HULL)						
	Gross α	Sb-125	Ru-106	Ce-137	Ce-134	Ce-136	Sr-90	Gross α	Sb-125	Ru-106	Ce-137	Ce-13·	Ce-144	Sr-90
9M HNO₃ - 3 x 6 hour contact at reflux														
C2/E	6.5	3559	59778	477	235	947	NM	0.09	–	15810	–	–	–	NM
C2/H	3.9	3707	30406	532	237	521	NM	0.25	–	14078	–	–	–	NM
C2/I	4.5	4966	30971	876	633	–	NM	0.17	–	13853	–	–	–	NM
C2/J	7.4	4133	41880	632	286	768	NM	0.08	–	12815	–	–	–	NM
A1/B	45.9	2664	62523	276	–	1667	NM	1.20	368	12817	146	·	–	NM
A1/C	20.2	1159	33685	227	–	1417	NM	1.40	–	8792	127	–	–	NM
A1/E	20.1	1630	51168	181	–	969	NM	0.22	–	10,029	64	–	–	NM
A1/F	13.4	1172	30381	134	–	2194	NM	0.25	190	8416	71	–	1516	NM
A3/E	6.4	458	12036	75	2	305	NM	0.16	–	1436	39	–	–	NM
A5/L	2.1	306	10593	16	–	114	NM	0.03	–	1262	–	–	–	NM
A5/Q	4.5	1119	39742	34	–	–	NM	0.05	–	1510	–	–	–	NM
9M HNO₃ - 3 x 6 hour contacts at 60°C														
A3/A	7.4	560	15944	65	3	79	NM	1.386	214	14187	50	–	–	NM
A3/B	4.5	435	10381	60	2	73	NM	0.724	159	8810	51	–	–	NM
9M HNO₃ - 3 x 2 hour contact at reflux														
C6/A	14.2	2337	22272	403	83	764	160	0.23	592	9571	194	–	–	34.2
C6/C	6.0	1908	23225	449	113	348	NM	0.33	235	12356	175	–	–	NM
A4/E	2.28	500	7613	442	59	230	NM	0.19	–	1184	29	–	–	NM
A4/G	6.69	750	16152	626	80	285	36	0.23	–	900	22	–	–	5.3
E1/1/C	75.2	1299	154756	338	135	20438	NM	0.22	117	12465	167	84	13261	NM
E1/1/F	148.1	2051	235594	418	148	20212	1674	0.18	84	6697	172	65	4515	77.9
E1/2/A	1.07	52	8708	19	1	–	NM	0.096	–	2599	17	–	–	NM
E1/2/C	1.12	57	6448	15	1	130	NM	0.110	–	4217	17	–	–	NM
E1/10/D	187.5	830	70610	347	89	7160	NM	0.62	251	26799	13	–	–	NM
D1/J	11.7	160	1893	21	4	42	NM	0.59	17	543	1	–	4	NM
9M HNO₃ - 3 x 1 hour contact at reflux														
C6/A	19.5	661	8957	199	16	390	NM	0.22	–	2705	86	–	–	NM
A3/F	3.8	417	10876	66	3	123	NM	0.33	–	5795	48	–	–	NM
E2/31A	209.6	4721	427842	394	96	23638	1122	3.6	453	60727	46	–	–	9.·
E2/31E	84.9	2121	232702	149	39	7923	NM	1.0	203	111123	–	–	–	NM
6M HNO₃ - 3 x 6 hour contact at reflux														
C6/E	6.1	2291	19077	543	79	885	NM	0.18	387	7809	339	–	–	NM
C6/G	4.9	1454	16384	498	95	54	167	0.14	–	6710	255	–	–	3.3
6M HNO₃, 3 x 2 hour contact at reflux														
C3/C	25.6	3718	79907	175	–	3029	NM	1.61	511	29452	105	–	1451	NM

NM - not measured.

Table 15 Decontamination of PFR Hulls by Nitric Acid Containing Ceric Ions

Hull Identification	Burn-up (%)	Mn-54:Co-60	PERCENTAGE REMOVALS							Weight Loss (%)	Calculated Thickness Removed (μm)
			Gross α	Sb-125	Ru-106	Cs-137	Cs-134	Ce-144	Sr-90		
0.1M Ce(IV) – 2M HNO$_3$ 1 hour contact at 92°C											
C1/H	7.3	1.48	99.1	100	61.1	100	100	100	99.97	10.3	12
C1/I	7.3	1.48	98.7	100	66.6	100	-	100	NM	7.5	9
C2/A	7.3	73.9	98.5	81.2	52.0	100	100	100	NM	9.8	10
C6/B	7.3	74.0	98.2	100	35.7	100	100	100	97.0	10.1	12
C6/D	7.3	74.0	96.3	100	4.2	100	100	100	NM	8.6	10
A4/A	5.6	11.2	96.2	100	67.6	92.8	100	100	NM	11.7	13
A4/F	5.6	11.5	97.7	100	41.1	94.2	100	100	92.4	10.6	12
E2/3/C	4.1	107.5	97.1	94.3	39.0	88.4	100	-	NM	8.0	11
E2/3/F	4.1	108.1	94.0	93.0	16.0	83.1	100	100	82.3	8.2	12
0.05M Ce(IV) – 2M HNO$_3$ 1 hour contact at 90-95°C											
C3/D	7.3	7.8	75.3	69.1	7.3	100	100	100	NM	5.4	6
C3/E	7.3	7.4	34.0	36.4	2.2	41.4	100	100	NM	4.8	5
0.05M Ce(IV) – 2M HNO$_3$ 1 hour contact at reflux											
C3/I	7.3	6.6	97.8	95.6	16.6	100	100	100	NM	8.6	10

NM – not measured.

Table 16   Activities on Hulls Before and After Decontamination with Nitric Acid in the Presence of Ceric Ions

Hull Identi-fication	ACTIVITY BEFORE DECONTAMINATION (μCi/g HULL)							ACTIVITY AFTER DECONTAMINATION (μCi/g HULL)						
	Gross α	Sb-125	Ru-106	Ce-137	Ce-134	Ce-144	Sr-90	Gross α	Sb-125	Ru-106	Ce-137	Ce-134	Ce-144	Sr-90
0.1M Ce(IV) – 2M HNO_3 1 hour contact 92°C														
C1/H	11.8	1192	2078	38	17	657	10.9	0.10	-	809	-	-	-	0.004
C1/I	9.8	1159	2636	28	-	585	NM	0.13	-	880	-	-	-	NM
C2/A	27.8	5112	58983	850	418	2788	NM	0.43	847	20025	-	-	-	NM
C6/B	9.9	1808	7571	1145	420	1443	21.2	0.18	-	4868	-	-	-	0.64
C6/D	6.0	1858	52324	1247	439	966	NM	0.22	-	50113	-	-	-	NM
A4/A	5.0	459	1556	681	81	292	NM	0.19	-	504	49	-	-	NM
A4/F	6.1	919	11665	691	88	429	29.5	0.14	-	6866	40	-	-	2.3
E2/3/C	116.2	2923	54023	215	80	-	NM	3.35	166	32943	25	-	-	NM
E2/3/F	100.1	3337	74920	225	61	10552	80.1	5.96	233	62930	38	-	-	14.2
0.05M Ce(IV) – 2M HNO_3 1 hour contact at 90-95°C														
C3/D	11.3	1973	43202	73	29	1060	NM	2.81	609	40043	-	-	-	NM
C3/E	14.7	2408	100204	128	21	777	NM	9.72	1532	98015	75	-	-	NM
0.05M Ce(IV) – 2M HNO_3 1 hour contact at reflux														
C3/I	18.12	2120	38151	106	38	1667	NM	0.39	94	31824	-	-	-	NM

NM – not measured.

Table 17   Decontamination of PWR Hulls by AP – CITROX

Hull Identi-fication	Burn-up (%)	Mn-54:Co-60	PERCENTAGE REMOVALS							Weight Loss (%)	Calculated Thickness Removed (μm)
			Gross α	Sb-125	Ru-106	Cs-137	Cs-134	Ce-144	Sr-90		
3% KMnO₄ – 10% NaOH followed by 2.5% H₂C₂O₄ – 5% DAHC*											
C1/A	7.3	1.53	98.4	100	97.1	100	–	100	NM	0.2	<0.5
C1/C	7.3	1.56	95.8	100	97.6	100	100	100	NM	0.3	<0.5
C1/D	7.3	1.49	96.3	100	98.0	100	100	100	NM	1.8	2
C1/E	7.3	1.49	97.5	100	98.2	100	–	100	NM	0.1	<0.5
A4/B	5.6	11.2	95.2	100	88.7	100	100	100	NM	2.2	3
A4/T	5.6	11.2	91.0	100	81.2	88.5	100	100	28.3	0.9	1
E1/1/A	4.1	69.1	93.9	66.8	46.0	5.3	4.3	1.4	0.7	0.7	1
E1/1/B	4.1	69.9	93.0	61.0	18.6	4.7	4.9	1.7	NM	0.2	<0.5
1.5% KMnO₄ – 5% NaOH followed by 2.5% H₂C₂O₄ – 5% DAHC											
C1/B	7.3	1.52	91.4	100	89.3	100	100	100	95.6	0.8	1
C6/F	7.3	1.45 –	89.8	100	64.4	100	100	–	NM	1.6	2

* DAHC – Diammonium hydrogen citrate.
NM – not measured.

Hull Identi- fication	ACTIVITY BEFORE DECONTAMINATION (μCi/g HULL)							ACTIVITY AFTER DECONTAMINATION (μCi/g HULL)						
	Gross α	Sb-125	Ru-106	Cs-137	Cs-134	Ce-144	Sr-90	Gross α	Sb-125	Ru-106	Cs-137	Cs-134	Ce-144	Sr-90
3% KMnO$_4$ - 10% NaOH followed by 2.5% H$_2$C$_2$O$_4$ - 5% DAHC*														
CI/A	6.4	972	12865	25	-	166	NM	0.11	-	366	-	-	-	NM
CI/C	3.4	1050	18584	21	1	18	NM	0.14	-	451	-	-	-	NM
CI/D	7.0	1162	23447	33	2	113	NM	0.26	-	465	-	-	-	NM
CI/E	8.0	1176	20009	34	-	128	NM	0.20	-	372	-	-	-	NM
A4/B	5.0	638	15594	696	85	34	NM	0.24	-	1760	-	-	-	NM
A4/I	3.6	507	11723	647	72	71	34.2	0.32	-	2207	74	-	-	24.5
EI/I/A	4.9	385	13808	264	69	11384	100.8	0.30	128	7450	250	66	11219	100.2
EI/I/B	4.0	395	29556	279	69	8671	NM	0.28	154	24042	266	66	8521	NM
1.5% KMnO$_4$ - 5% NaOH followed by 2.5% H$_2$C$_2$O$_4$ - 5% DAHC														
CI/B	5.0	1116	13543	23	11	171	8.1	0.43	-	1464	-	-	-	0.36
C6/F	4.6	2201	27884	1133	422	-	NM	0.47	-	9916	-	-	-	NM

*DAHC - Diammonium hydrogen citrate.
NM - not measured.

Table 19   Details of Typical AP-CITROX Decontaminations

% ACTIVITY REMOVALS

Hull Cl/E	Gross α	Fission Products					Activation Products		
		Sb-125	Ru-106	Cs-137	Cs-134	Ce-144	Mn-54	Co-60	Co-
AP	79.6	89.9	95.2	97.1	–	0	0.6	0.2	
CITROX	17.9	10.1	3.0	2.9	–	100	0.3	0.4	0.
TOTAL	97.5	100	98.2	100	–	100	0.9	0.6	0.
Hull Cl/B									
AP	54.7	82.7	83.7	95.7	90.0	0	0.3	0	
CITROX	36.7	17.3	5.6	4.3	9.1	100	0.4	0.3	0.
TOTAL	91.4	100	89.3	100	100	100	0.7	0.3	0..

Table 20 Decontamination of PWR Rods with Oxalic Acid

Rod Identification	Burn-up (GWD)	Mn-54/Co-60	CONTACTING Duration (hr)	Number	Gross α	Sb-125	PERCENTAGE REMOVALS Ru-106	Co-137	Co-134	Co-144	β-90	Weight Loss (%)	Calculated Thickness Removed (μm)
0.5M Oxalic Acid													
C1/F	7.3	1.3	6	1	85.1	64.0	15.7	100	100	100	91.1	3.1	?
C6/I	7.3	75.1	6	2	89.1	90.4	38.3	40.8	36.6	100	NM	9.0	11
A4/D	3.6	113	6	2	86.0	91.6	82.2	96.3	100	100	NM	12.3	15
A4/B	3.6	11.2	6	2	36.6	79.8	35.3	68.4	45.9	100	95.1	9.0	11
0.2M Oxalic Acid – 1M H₂O₂													
C1/G	7.3	1.3	6	1	98.3	91.1	69.0	100	-	100		0.3	-
C6/H	7.3	75.3	6	2	96.7	100	37.7	42.0	100	??	53.0	2.4	-
C6/J	7.3	28.0	6	2	96.7	100	36.6	45.7	100	100	NM	2.3	2
A4/C	3.6	11.2	6	2	90.7	100	66.3	90.6	100	100	NM	2.3	2
A4/J	3.6	11.2	6	2	8?	18?	39.2	98.6	100	21.3	NM	1.9	?
A3/C	3.6	11.4	6	2	97.7	94.7	36.8	24.2	100	28.3	16.3	0.7	-
A3/G	3.6	10.2	2	2	99.00	100	61.9	27.4	100	64.0	NM	1.0	-
B1/7/B	4.1	69.3	2	2	99.93	53.3	94.6	47.7	45.0	60.7	46.0	1.7	?
B1/11/E	4.1	70.2	1	2	33.9	53.7	96.3	56.6	67.3	100	NM	1.4	2
B1/C	0	2.3	2	2	99.44	95.1	79.1	98.5	100	100	NM	0.9	-
0.2M Oxalic Acid – 1M H₂O₂													
A3/E	3.6	11.0	2	2	97.2	100	51.5	33.0	-	100	NM	1.3	2
0.1M Oxalic Acid – 1M H₂O₂													
C6/D	7.3	11.7	2	2	99.12	100	62.7	29.9	100	100	22.0	2.1	2
A3/I	3.6	9.6	2	2	95.0	100	30.8	19.3	100	100	NM	0.8	1
B1/10/C	4.1	2.6	2	2	62.0	53.6	26.3	96.8	100	100	NM	0.9	0
B1/12/C	4.1	2.3	2	2	90.4	100	100	96.5	100	100	NM	1.7	0

- isotope not detected
NM - not measured
N/A - not applicable
* not calculated - different geometry of ring.

Table 21   Activities on Hulls Before and After Decontamination with Oxalic Acid

Hull Identification	ACTIVITY BEFORE DECONTAMINATION (μCi/g HULL)							ACTIVITY AFTER DECONTAMINATION (μCi/g HULL)						
	Gross α	Sb-125	Ru-106	Cs-137	Cs-134	Ce-144	Sr-90	Gross α	Sb-125	Ru-106	Cs-137	Cs-134	Ce-144	Sr-90
0.5M Oxalic Acid														
C1/F	8.1	1558	20030	29	12	331	16.0	1.2	361	16887	–	–	–	-1.1
C6/I	4.2	2354	34647	500	159	1410	NM	0.25	226	14443	296	65	–	NM
A4/D	1.3	440	4815	320	36	131	NM	0.17	37	856	44	–	–	NM
A4/H	3.6	1423	29435	434	122	321	40.8	1.54	287	13155	137	66	–	2.0
0.5M Oxalic Acid – 1M H₂O₂														
C1/G	13.4	1974	28577	31	–	1160	NM	0.24	176	8854	–	–	–	NM
C6/H	4.4	1612	18906	496	77	249	8.6	0.14	–	11782	282	–	–	4.0
C6/J	3.7	1495	14434	446	75	–	NM	0.12	–	9155	252	–	–	NM
A4/C	2.9	603	9397	468	52	21	NM	0.27	–	2904	95	–	–	NM
A4/J	2.4	420	4119	393	28	82	NM	0.23	–	1675	194	–	–	NM
A3/C	4.7	637	16274	64	3	403	NM	0.17	34	7031	90	–	309	NM
A3/L	5.7	535	12867	62	4	410	30.9	0.13	–	4892	45	–	293	25.9
E1/1/D	159.8	1723	209418	426	173	10267	NM	0.13	133	7102	223	87	6427	NM
E1/1/E	199.9	2148	275952	479	138	38575	172.5	0.13	134	9731	208	45	15353	93.1
E1/12/D	41.4	44	1524	216	721	1476	NM	19.1	–	–	–	13	–	NM
D1/C	32.9	286	3595	65	8	54	NM	0.18	14	746	–	–	–	NM
0.2M Oxalic Acid – 1M H₂O₂														
A3/D	6.8	619	16955	65	–	10	NM	0.19	–	8222	50	–	–	NM
0.1M Oxalic Acid – 1M H₂O₂														
C4/D	19.8	610	12557	184	10	281	93.9	0.18	–	4693	129	–	–	73.2
A3/1	4.4	456	10081	62	3	82	NM	0.18	–	6969	50	–	–	NM
E1/:0/C	146.2	672	73842	405	85	2667	NM	55.5	312	47037	13	–	–	NM
E1/12/C	41.7	47	1511	218	746	1532	NM	4.03	–	–	12	–	–	NM

NM – not measured.

Table 22 Decontamination of Mullo with LOMI Reagents

Bell No.	COMPOSITION OF LOMI REAGENT			CONTACT		PERCENTAGE REMOVALS						Weight Loss (%)
	$[V^{2+}]$	$[HCOOH]$	$[Picolinic Acid]$	Duration	Temperature	Gross γ	Sb125	Ru106	Co117	Co118	Co60	
C4/C	$6.8 \times 10^{-2}M$	$7.8 \times 10^{-2}M$	$4.25 \times 10^{-2}M$	24 hours	80-85°C	86.0	5.7	9.1	17.9	25.0	100	1.3
C4/R	$5.3 \times 10^{-2}M$	$6.43 \times 10^{-2}M$	$3.33 \times 10^{-2}M$	6 hours	80-85°C	87.6	5.8	10.3	19.8	100	100	1.1
E3/A/D		—	—	3 hours	Reflux	83.3	16.5	7.9	3.4	3.3	100	0.1
E3/A/B	$-6 \times 10^{-2}M$	$2.21 \times 10^{-1}M$	$1.63 \times 10^{-1}M$	6 hours	80-85°C	46.5	20.3	61.7	2.3	3.4	—	1.5
E3/A/A				3 hours	Reflux	47.5	27.3	56.6	2.9	6.3	0	0.3

Table 23   Hulls Used for Whole Hull Decontaminations

Hull Identification	Length (mm)	Radiation at 75 mm (rad/h)	Appearance Initially
E2A/2	25	5-10	Dull, black
E2A/4	50	5	Dull, black
E3A/4	50	2	Glossy, black
E3A/5	50	5-10	Shiny with large areas of black deposit

Table 24   Decontamination of Complete Hulls - Summary

Sample No.	Mn-54:Co-60	% REMOVALS								Wt.Loss (%)
		Gross α	Sb-125	Ru-106	Cs-137	Cs-134	Ce-144	Sr-90	Ru-155	
E3A/5	1.37	98.1	97.5	91.8	98.9	>99	-	93.6	>99	1.21
E2A/4	65.9	96.0	94.5	82.6	95.7	93.4	-	78.7	>99	1.98
E2A/2	96.4	98.6	97.5	95.5	99.0	98.8	-	97.0	>99	5.99
E3A/4	74.8	96.7	99.3	78.4	82.1	80.4	-	72.4	>99	2.69

Total Activity Before and After Decontamination

Sample No.	INITIAL ACTIVITY (μCi/g)						RESIDUAL ACTIVITY (μCi/g)					
	Gross α	Sb-125	Ru-106	Cs-137	Cs-134	Sr-90	Gross α	Sr-125	Ru-106	Cs-137	Cs-134	Sr-90
E3A/5	52.3	1510	193000	92	31	42	1.0	39	15894	1	< 0.3	3
E2A/4	44.2	1880	188000	234	72	42	1.8	118	33436	10	5	9
E2A/2	434.2	7690	1450000	1270	322	330	6.4	216	69161	17	4	11
E3A/4	7.5	1000	16000	108	30	60	0.26	8	.3557	20	6	17

Table 25   Decontamination of Complete Balls - Details

Sample No.	Contacting No.	Mn:Co	% REMOVALS						Visual Observation
			Gross α	Sb-125	Ru-106	Cs-137	Cs-134	Sr-90	
R2A/2		96.4							Initially matt, black.
	1		95.17	91.5	88.8	94.4	95.5	91.7	No change in appearance, but fine black particulates in the solution.
	2		2.56	4.1	5.0	3.7	3.7	3.7	)
	3		0.90	1.5	1.9	1.1	0.8	1.7	)No change in appearance)
	TOTAL		98.63	97.1	95.7	99.2	100	97.1	
R2A/4		68.9							Initially matt black.
	1		74.59	73.0	30.4	79.2	78.'	53.5	Becoming glossy in places due to removal of deposit
	2		18.99	19.5	48.5	13.7	9.3	18.3	)
	3		2.43	2.0	3.9	2.9	5.5	6.9	)No further change)
	TOTAL		96.01	94.5	82.6	95.8	93.2	78.7	
R3A/4		74.8							Initially glossy black.
	1		90.7	93.7	45.6	50.6	47.4	36.1	Becoming matt - probably due to pitting
	2		4.3	4.5	19.2	19.7	20.6	21.7	)
	3		1.7	1.3	13.6	11.7	12.3	14.4	)No further change)
	TOTAL		96.7	99.5	78.4	82.0	80.3	72.2	
R3A/5		1.37							Initially shiny with large areas of blk deposit.
	1		80.76	78.8	51.0	77.0	73.7	72.3	Much of deposit cleaned off. Greyish appearance where removal incomplete.
	2		15.42	16.9	37.3	17.7	19.3	17.3	Remaining deposit removed. leaving ball clean and shiny.
	3		1.88	1.7	3.6	4.1	7.0	4.1	No further change.
	TOTAL		98.06	97.4	91.7	98.8	100	93.7	

Table 26   Relative Activities Associated with Particulates
in Decontamination Solutions

	Sb-125	Ru-106	Cs-137
Particulates in Decontamination Solutions	$2 \times 10^{-3} - 2 \times 10^{-2}$	1	$2 \times 10^{-4} - 9 \times 10^{-3}$
Typical Dissolver Insolubles	$4 \times 10^{-3} - 6 \times 10^{-3}$	1	$4 \times 10^{-4} - 2 \times 10^{-3}$

TOP END CAP
FUEL RETAINER

UPPER AXIAL BREEDER

CORE

SPACER

LOWER
AXIAL
BREEDER

SPACER

GAS
PLENUM

END CAP
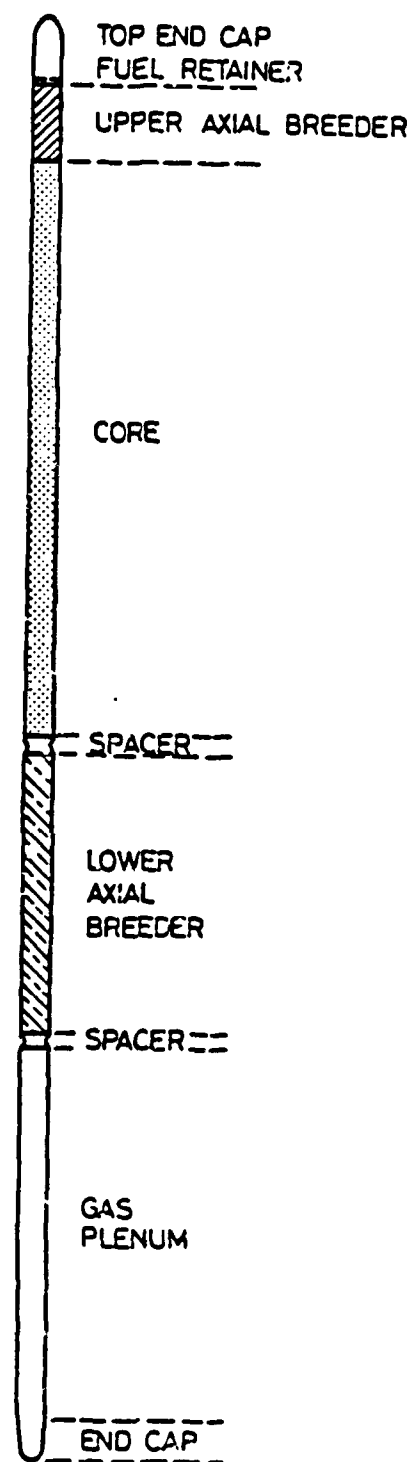
FIG. 1 DIAGRAMMATIC REPRESENTATION OF A PFR FUEL PIN
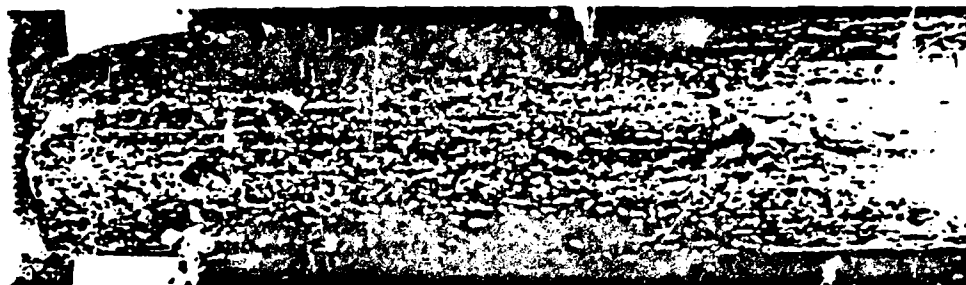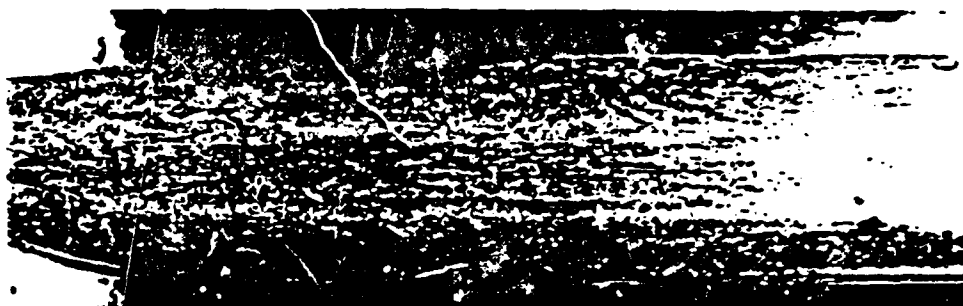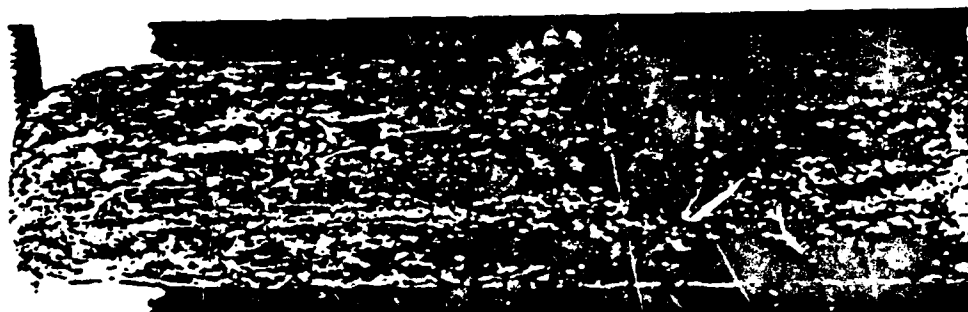
Fig. 2
Side views of some PFR hulls

(a) clean shiny hull
(b) crimped section at location of spacer
(c) hull with oxide film
(d) hull with oxide film and particulates

PFR hulls   Batch E/1

x 3.5
End 2

End 1

No. 1

No. 2

No. 3

No. 4
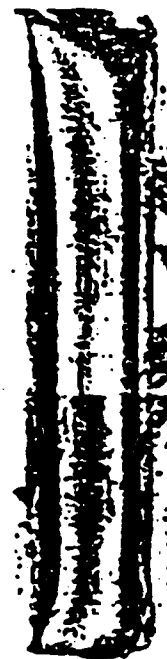
Fig. 3

PFR hulls   Batch A   x 3.5

End 1   No. 1   No. 2   No. 3   No. 4   End 2

Fig. 4
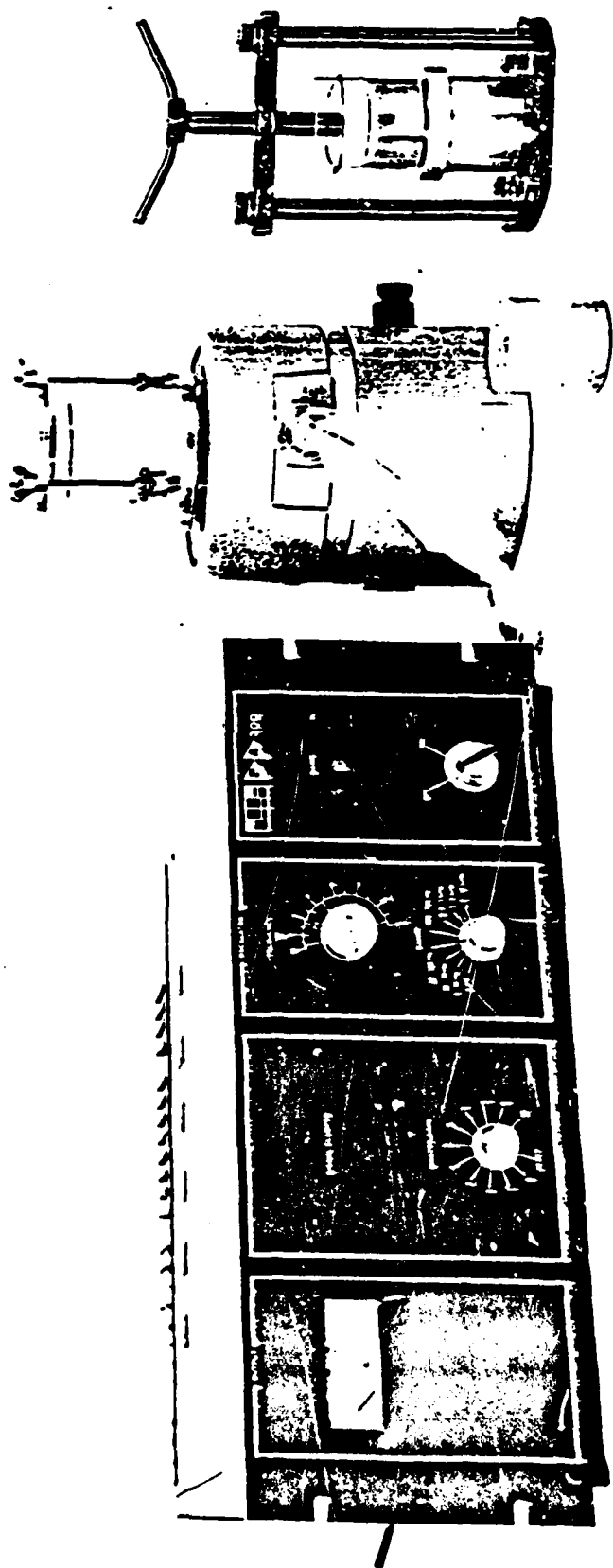
Fig. 5

Inactive equipment for the production of cement samples

Fig. 6
Stainless steel hulls incorporated in sand/OPC/water
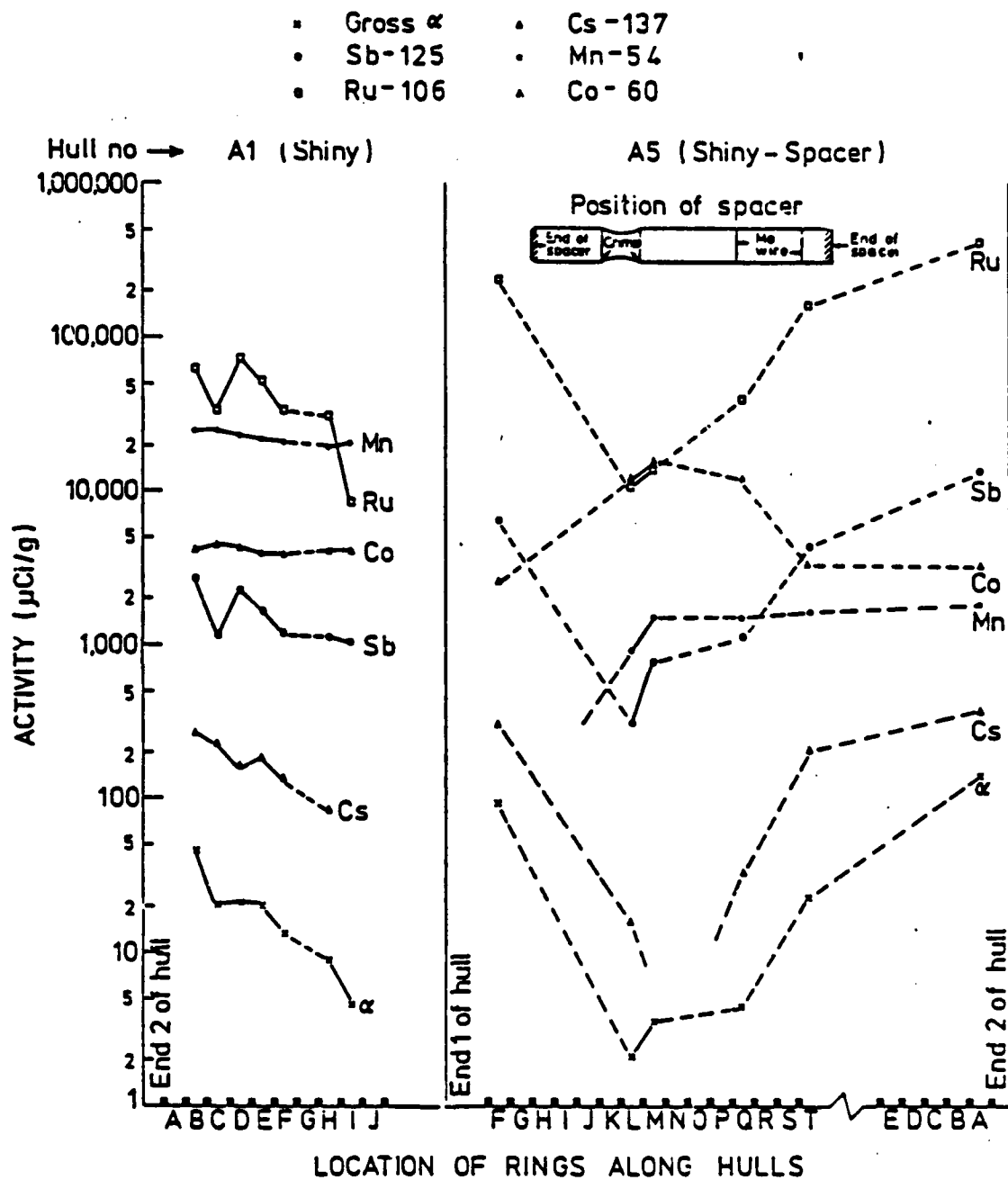
FIG. 7. PROFILES OF ACTIVITIES ACROSS 5·6% BURN UP HULLS (BATCH A)
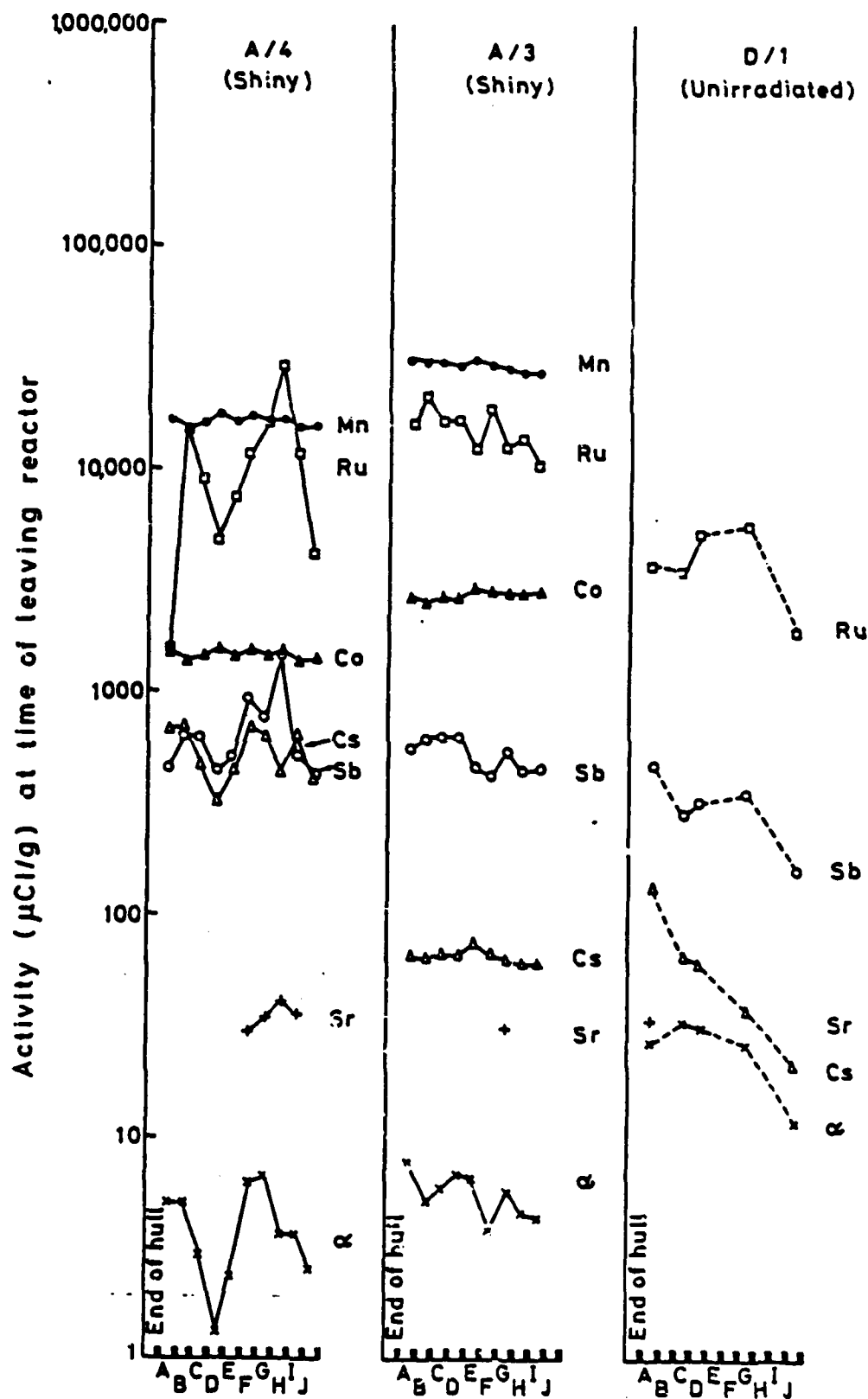
FIG. 8. PROFILES OF ACTIVITIES ACROSS 5·6 % BURN-UP
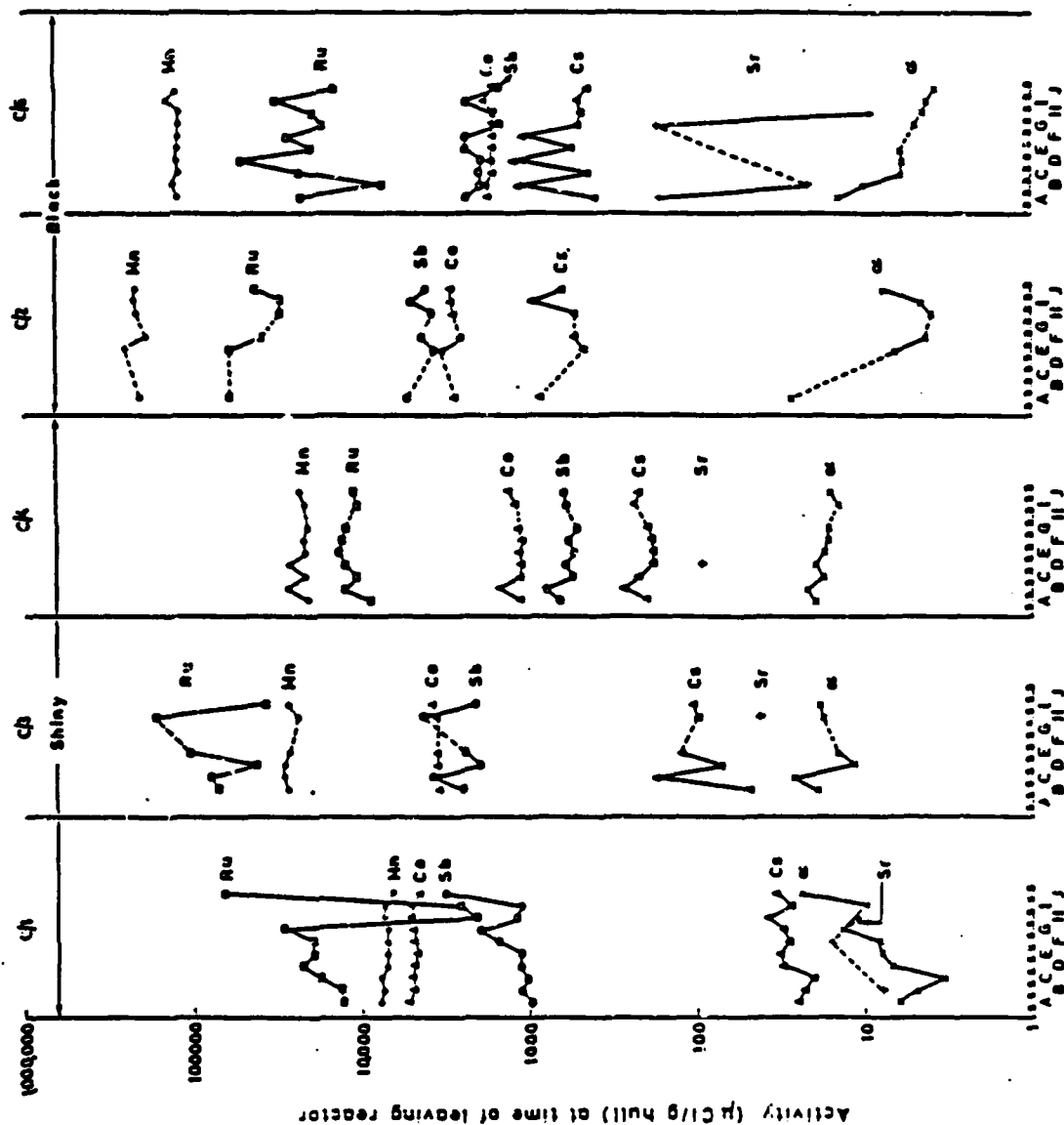
FIG. 9. PROFILES OF ACTIVITIES ACROSS 7.3% BURN-UP HULLS (BATCH C)
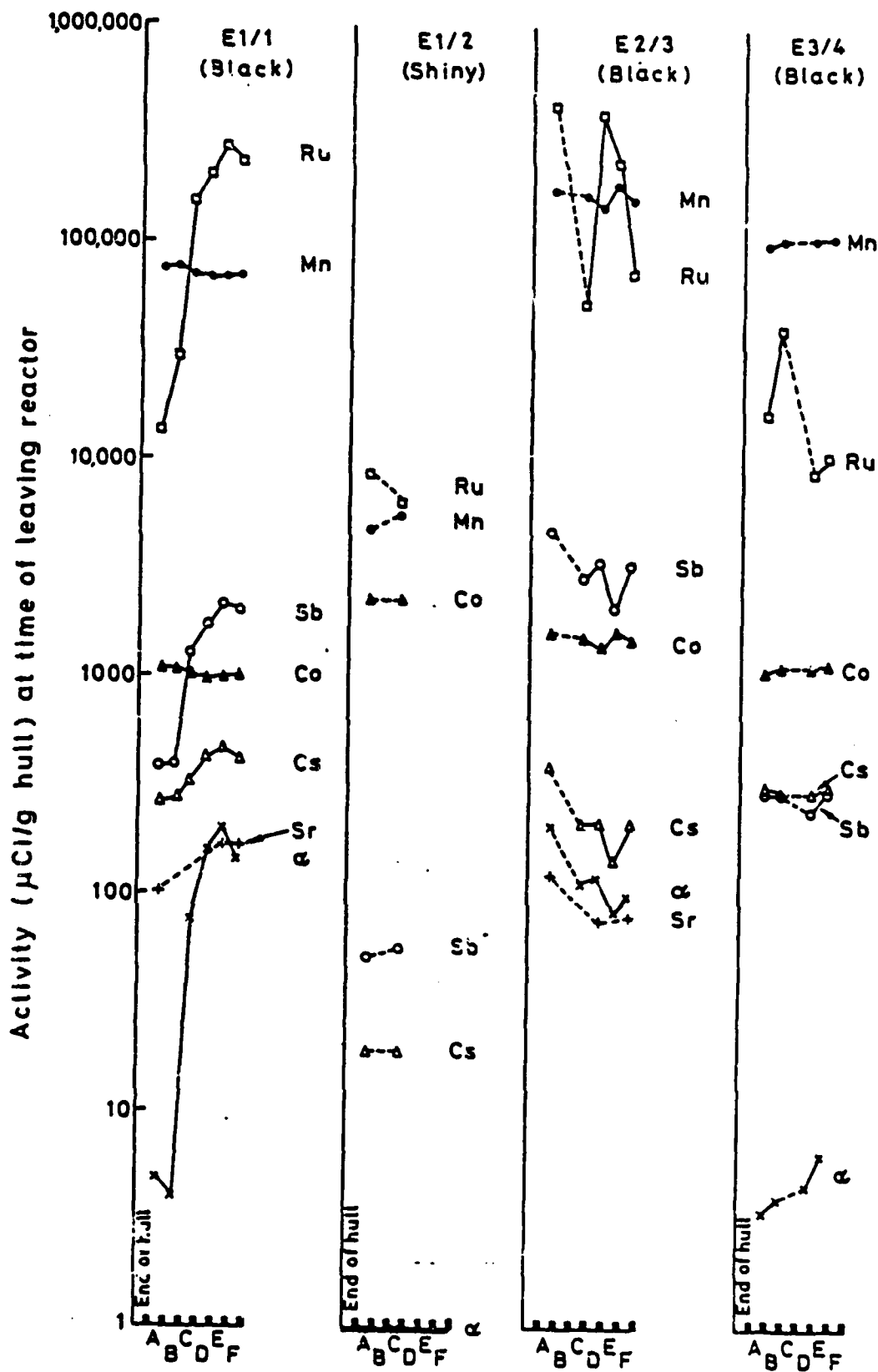
FIG.10. PROFILES OF ACTIVITIES ACROSS 4·1% BURN-UP HULLS
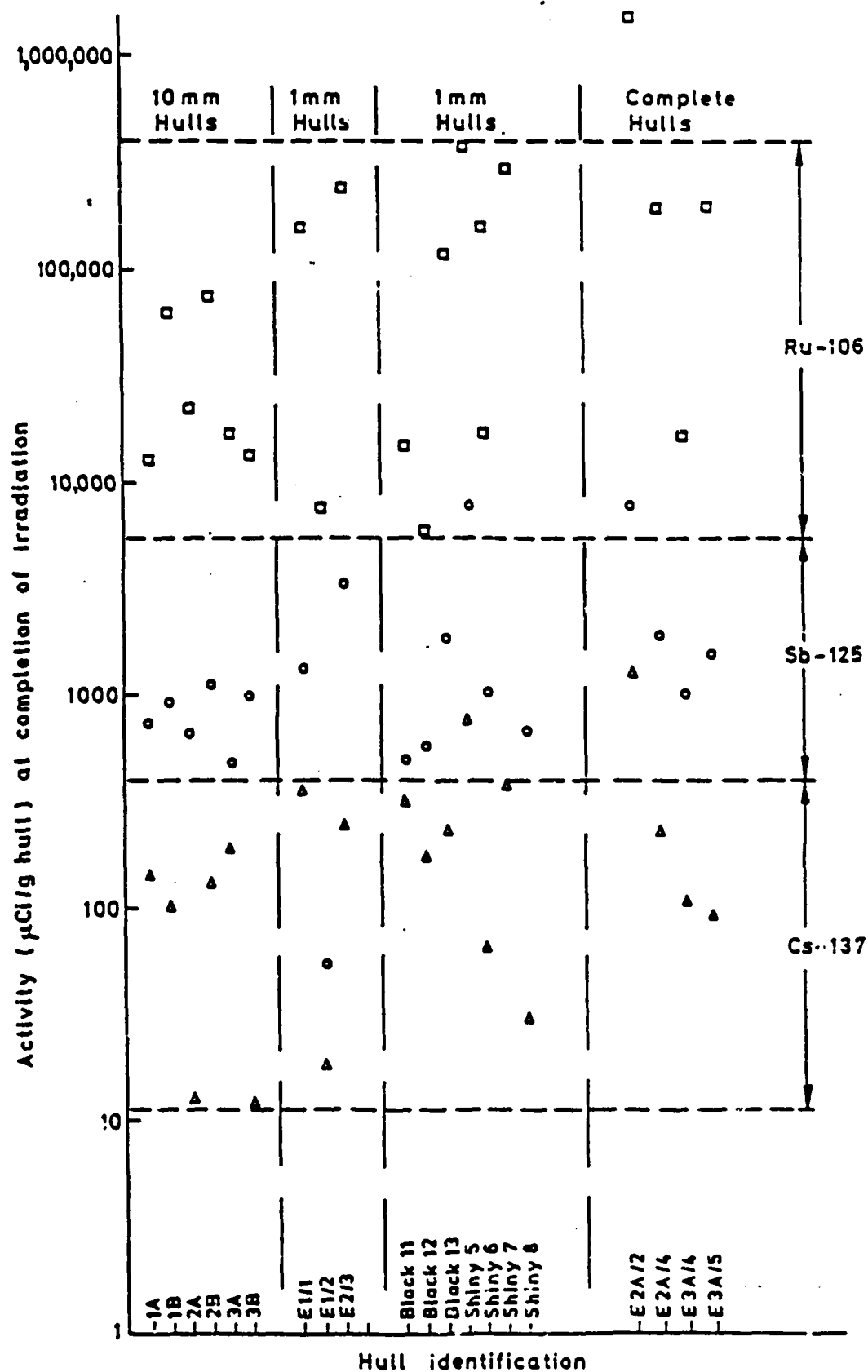
FIG.11. COMPARISON OF FISSION PRODUCT ACTIVITIES ON BATCH E HULLS.
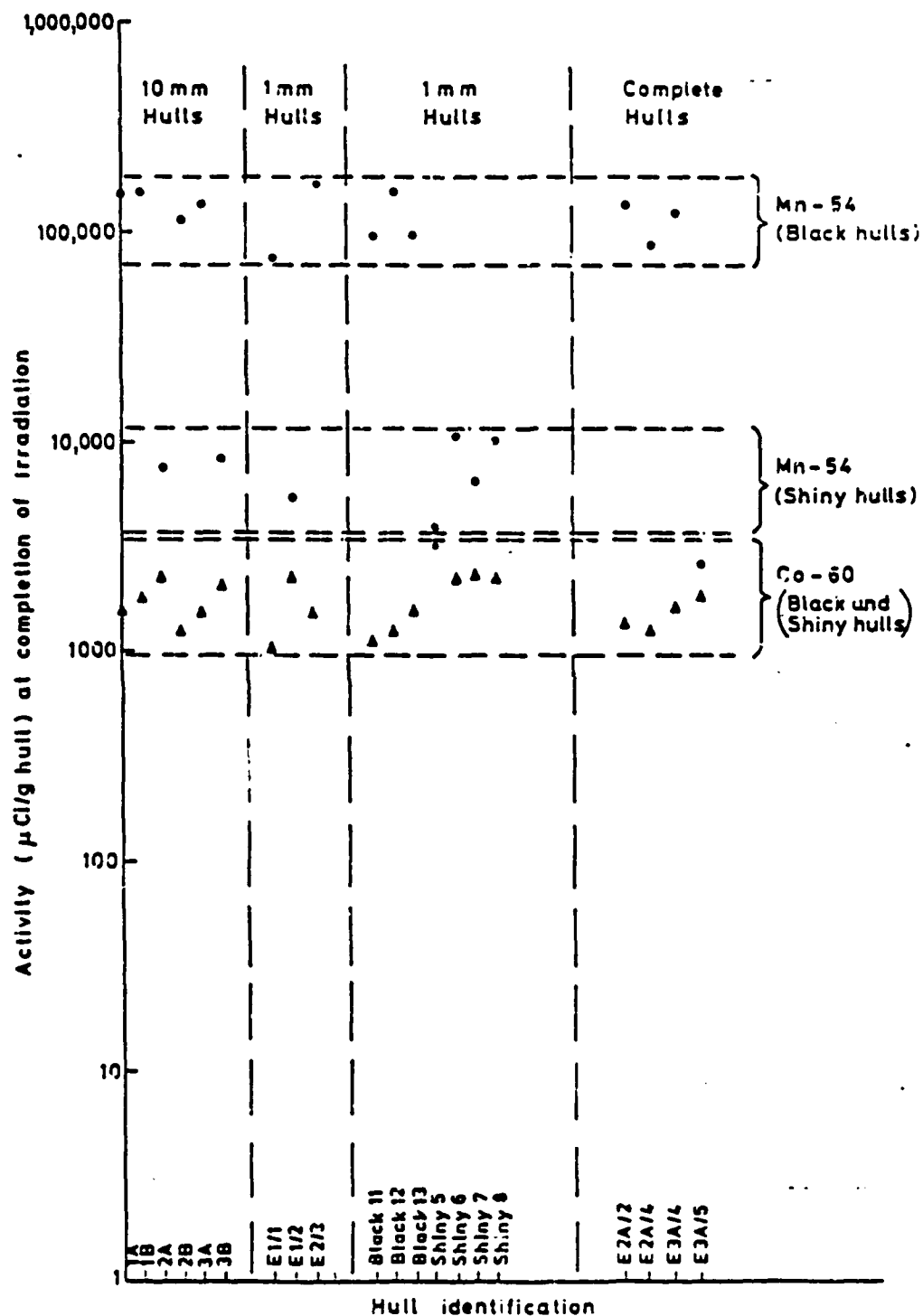
FIG.12. COMPARISON OF ACTIVATION PRODUCT ACTIVITIES ON BATCH
E HULLS.

E1 / 10
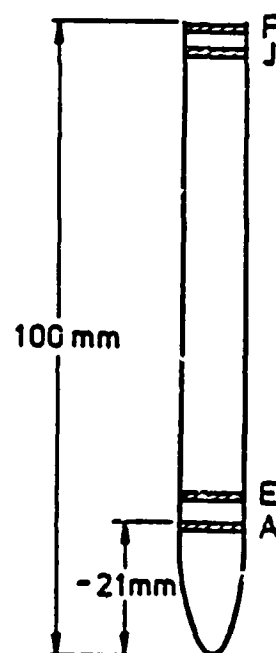
E2 / 4
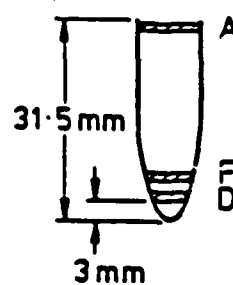
FIG. 13. LOCATION OF RINGS CUT
FROM END PIECE HULLS
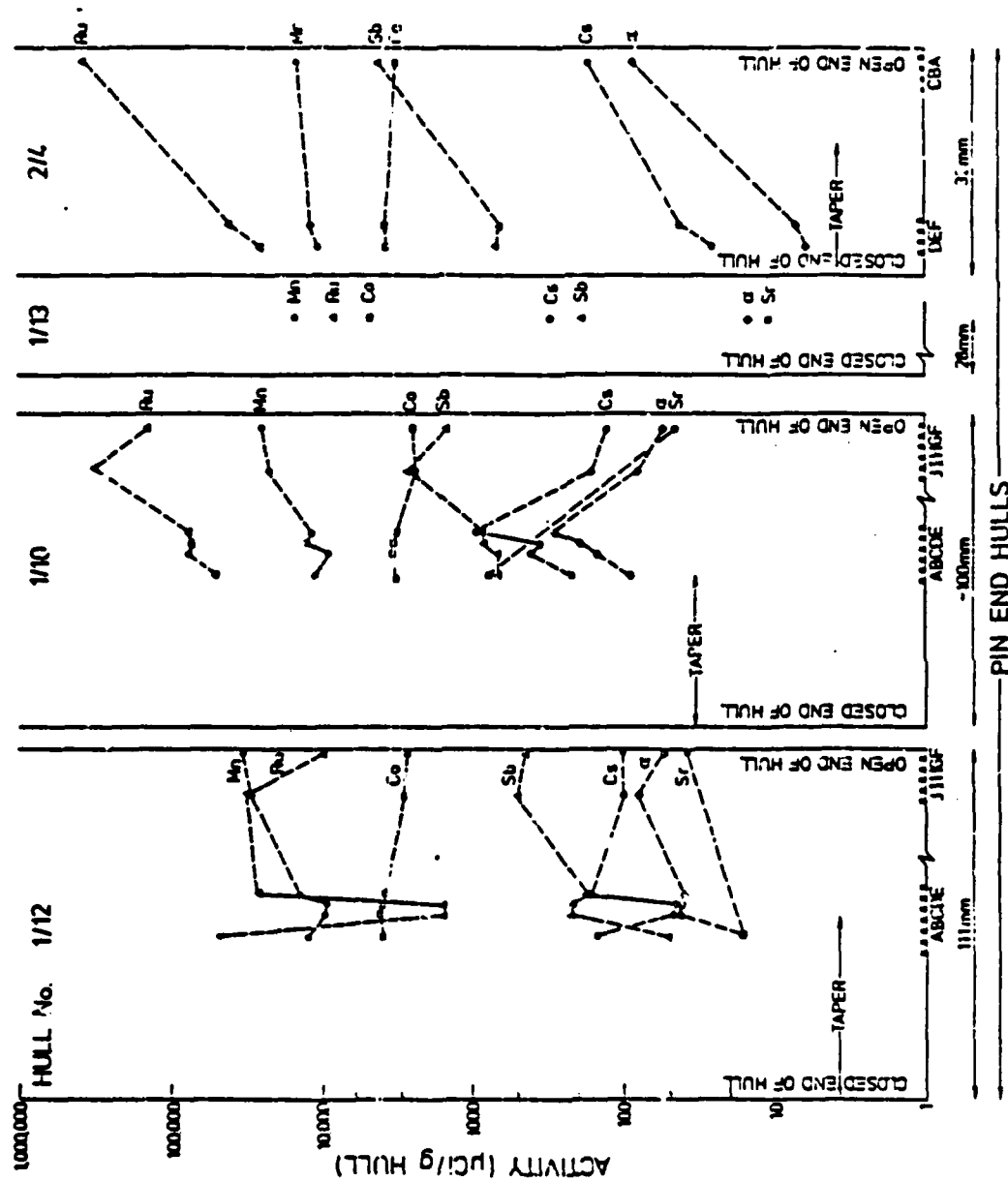
FIG. 14. ACTIVITY PROFILES OF END PIECE HULLS
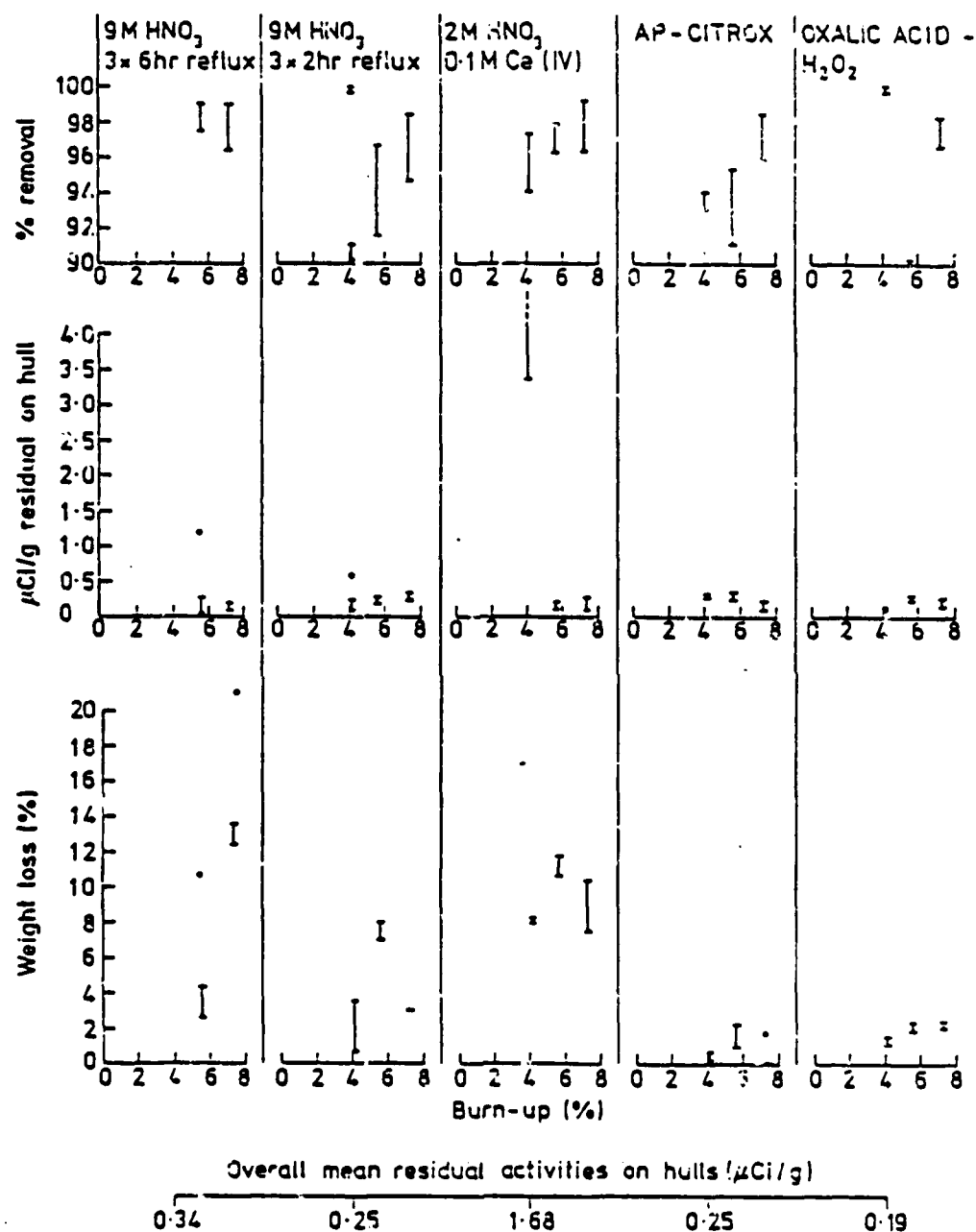
FIG. 15. COMPARISON OF THE DECONTAMINATION ROUTES IN
TERMS OF PERCENT α ACTIVITY REMOVALS AND
RESIDUAL α ACTIVITY ON HULLS

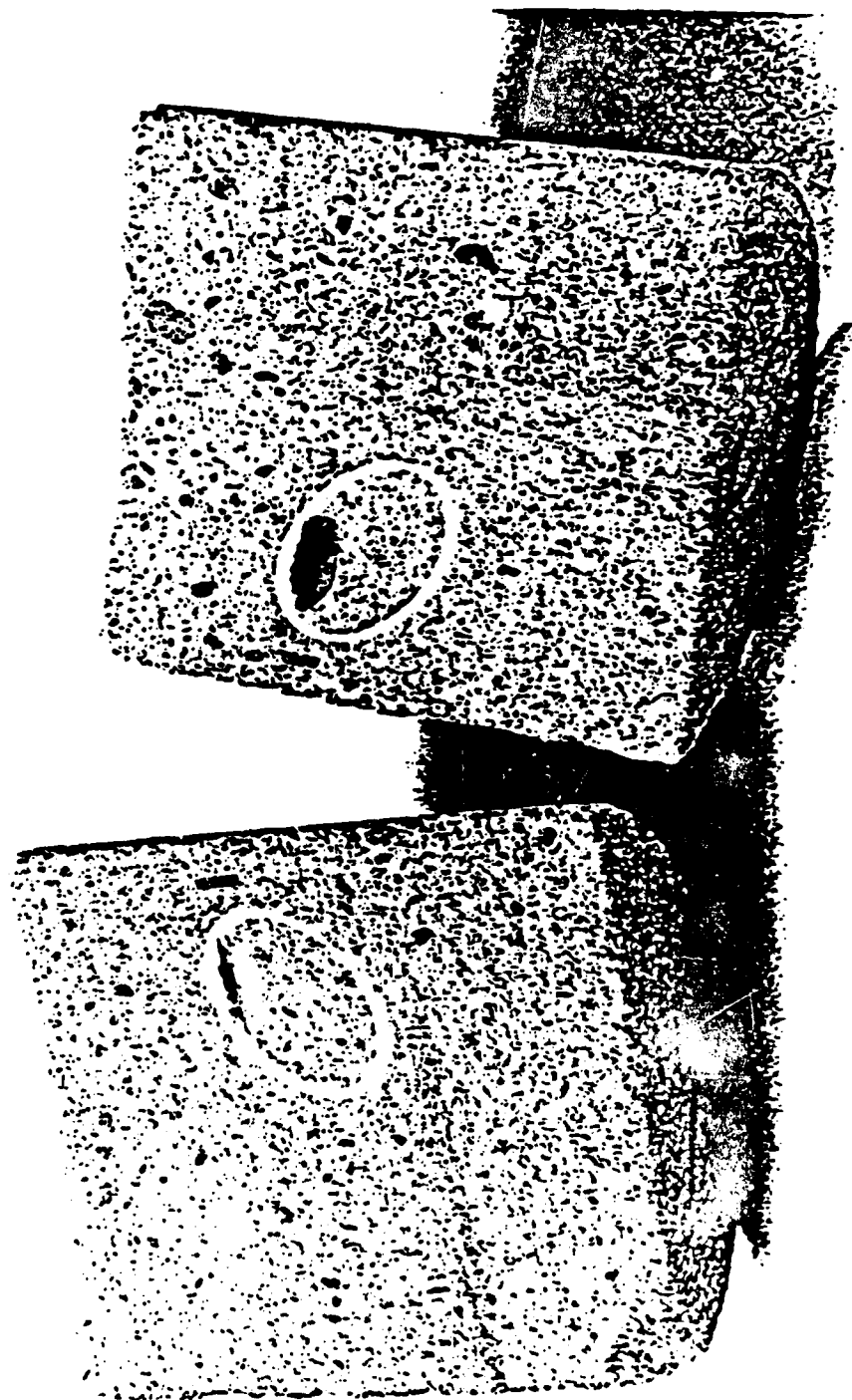FIG.16 HULL E3A/4 BEFORE (top) and AFTER (bottom) BEING

DECONTAMINATED WITH 9M HNO$_3$

Fig. 17
Voids in cemented samples

Fig. 18
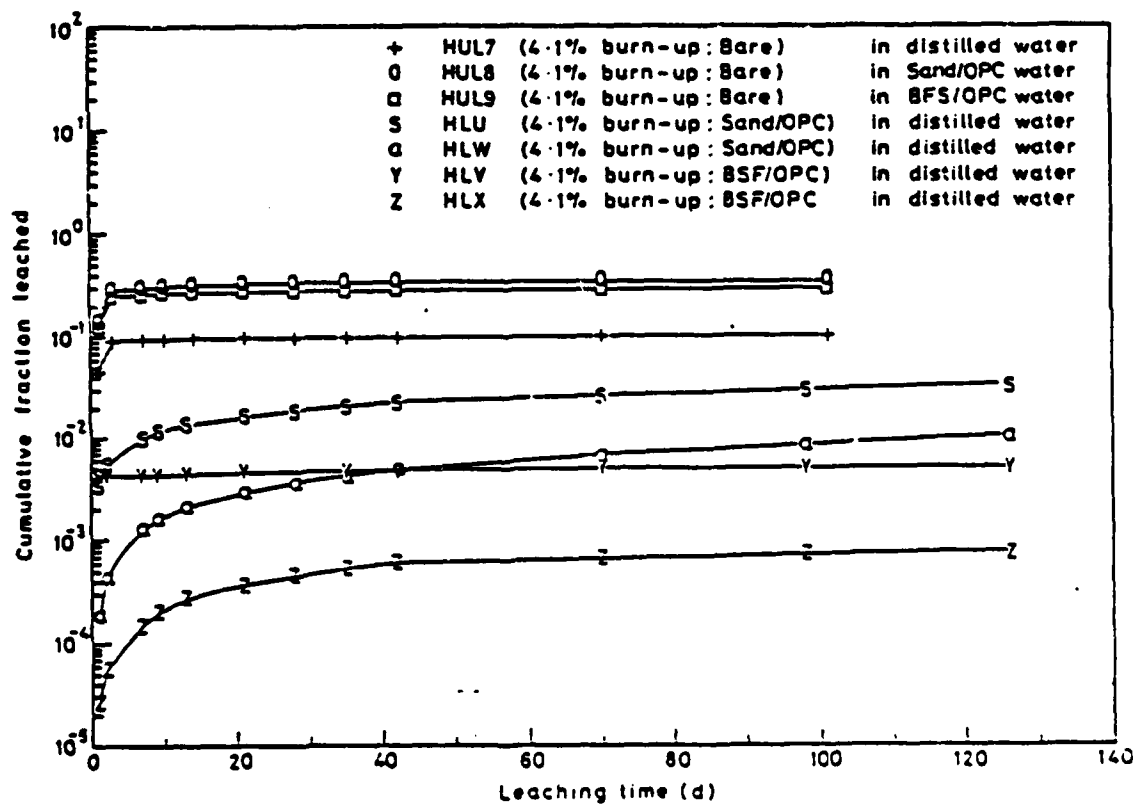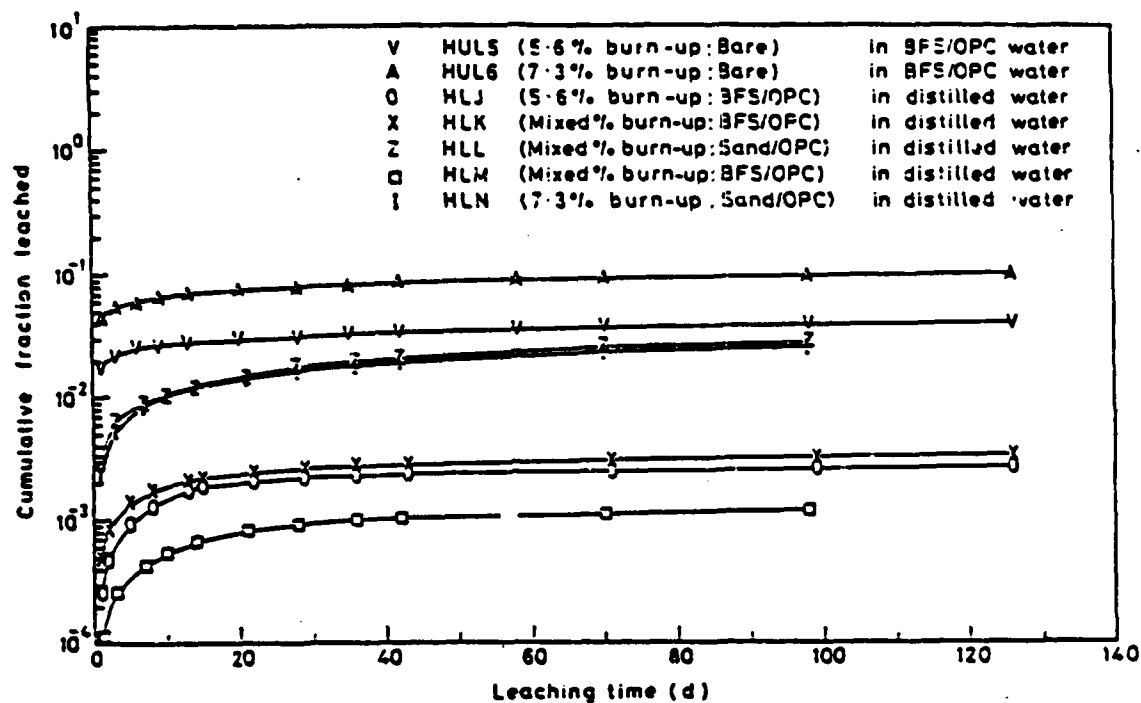Stainless steel hulls immersed in.
top sand OPC bottom BFS OPC

V	HUL5	(5·6% burn-up: Bare)	in BFS/OPC water
A	HUL6	(7·3% burn-up: Bare)	in BFS/OPC water
0	HLJ	(5·6% burn-up: BFS/OPC)	in distilled water
X	HLK	(Mixed% burn-up: BFS/OPC)	in distilled water
Z	HLL	(Mixed% burn-up: Sand/OPC)	in distilled water
□	HLM	(Mixed% burn-up: BFS/OPC)	in distilled water
I	HLN	(7·3% burn-up. Sand/OPC)	in distilled water

+	HUL7	(4·1% burn-up: Bare)	in distilled water
0	HUL8	(4·1% burn-up: Bare)	in Sand/OPC water
□	HUL9	(4·1% burn-up: Bare)	in BFS/OPC water
S	HLU	(4·1% burn-up: Sand/OPC)	in distilled water
a	HLW	(4·1% burn-up: Sand/OPC)	in distilled water
Y	HLV	(4·1% burn-up: BSF/OPC)	in distilled water
Z	HLX	(4·1% burn-up: BSF/OPC	in distilled water

Fig.19 Cumulative fraction of Sr-90 leached from bare and immobilised PFR Hulls
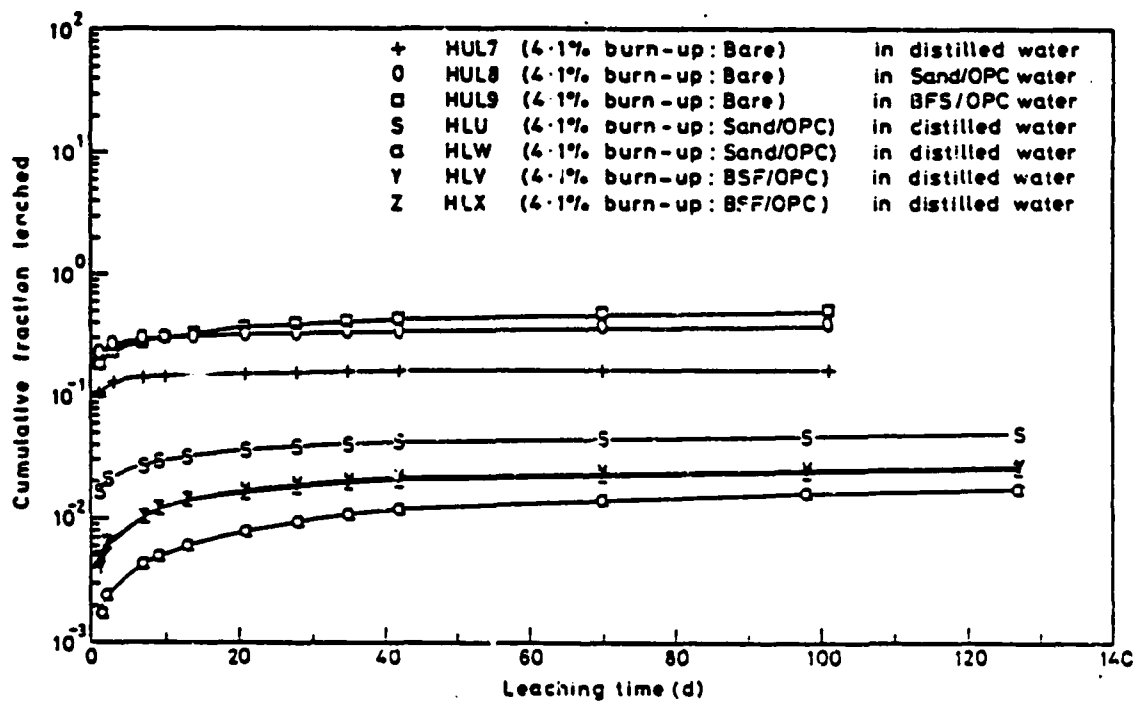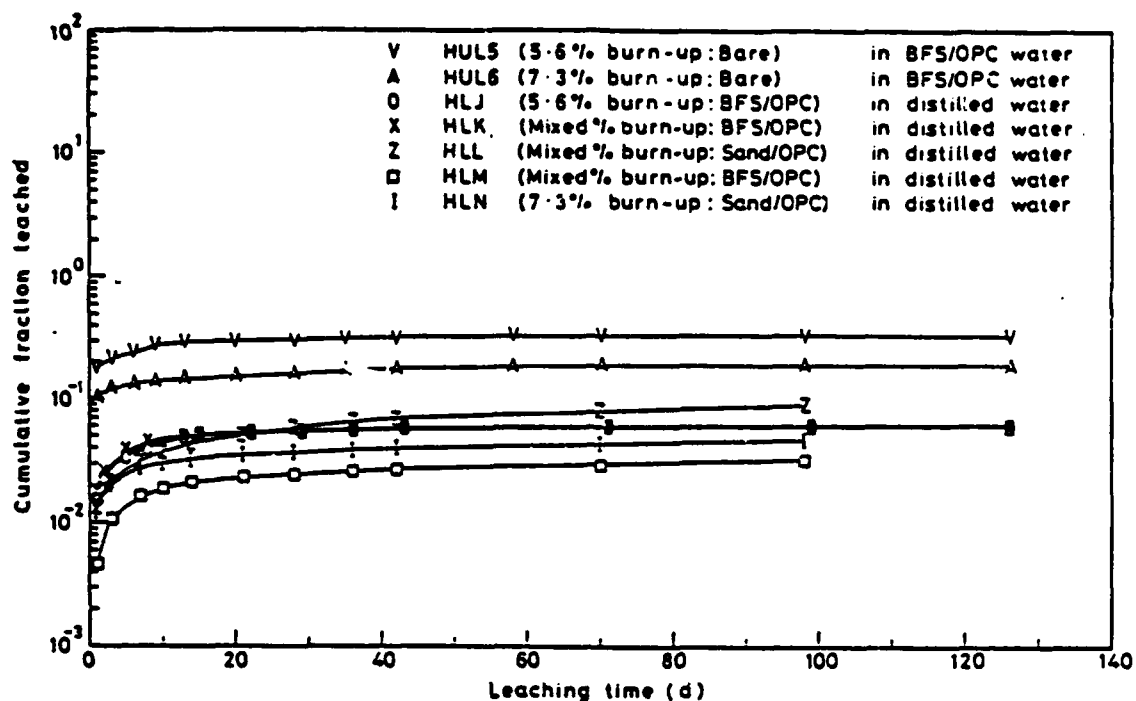
Fig. 20 Cumulative fraction of Cs-137 leached from bare and immobilised PFR Hulls.
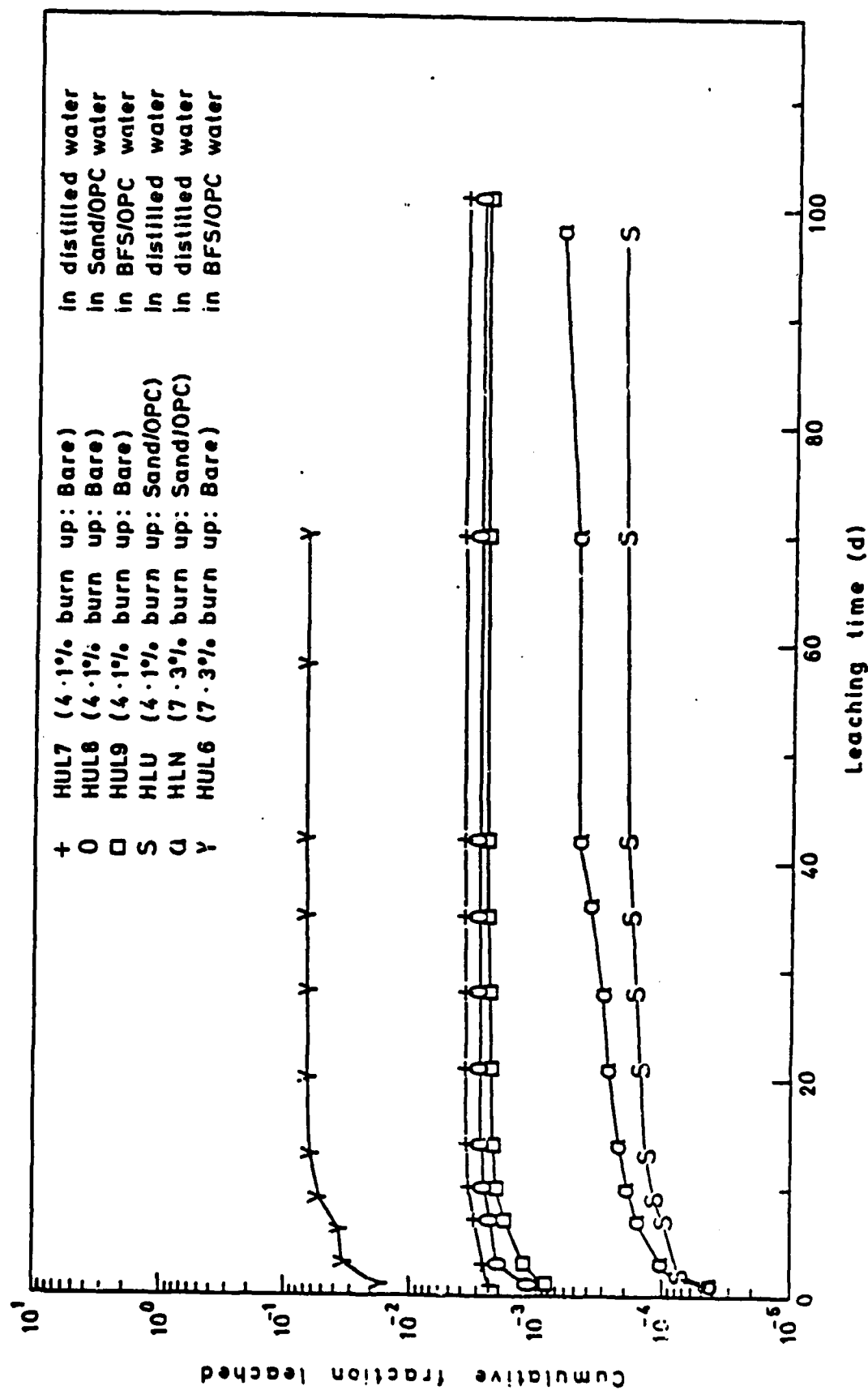
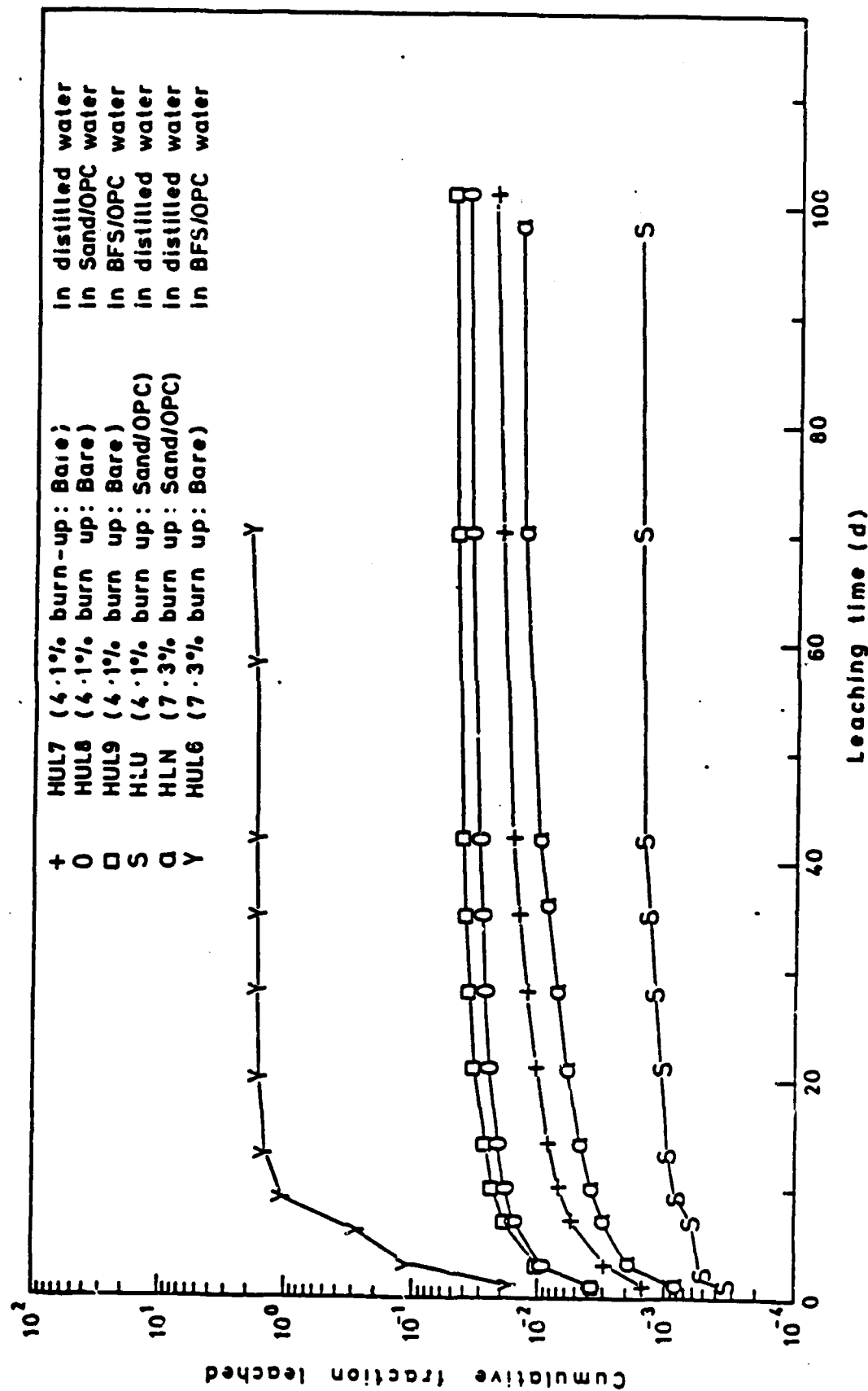Fig. 21 Cumulative fraction of Ru-106 leached from bare and immobilised PFR Hulls.

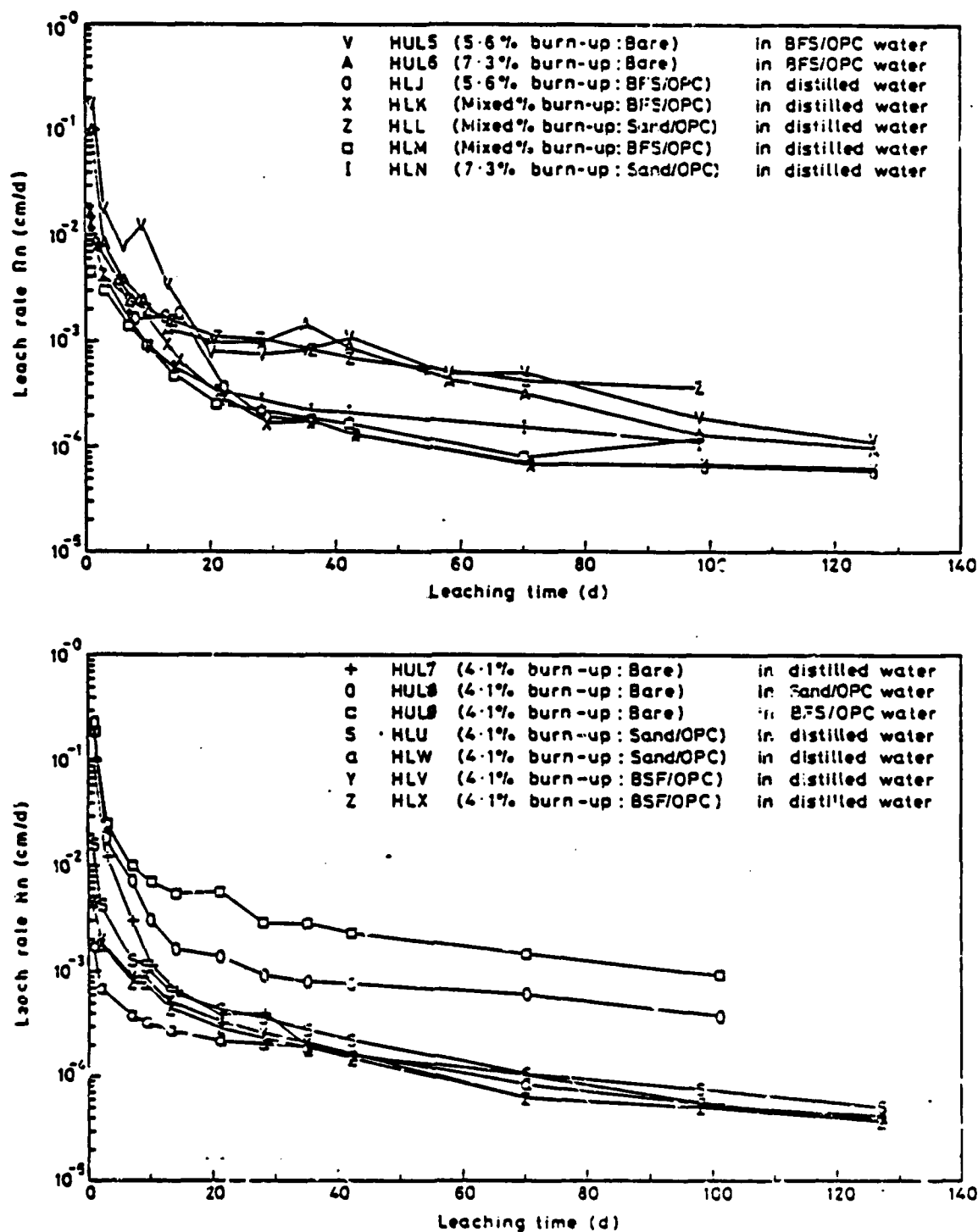Fig. 22 Cumulative fraction of Sb-125 leached from bare and immobilised PFR Hulls.

Fig. 23 Leach rates for Cs-137 from bare and immobilised PFR Hulls.

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